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Although this document is principally concerned with the health and welfare effects of lead, other scientific data are presented and evaluated in order to provide a better understanding of this pollutant in the environment. To this end, the document includes chapters that discuss: the chemistry and physics of the pollutant; analytical levels; atmospheric chemistry and dispersion modeling; effects on vegetation; and respiratory, physiological, toxicological, clinical and epidemiological aspects of human exposure.

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Research Triangle Park, NC 27711

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LIST OF ABBREVIATIONS

AAS	Atomic absorption spectrometry
Ach	Acetylcholine
ACTH	Adrenocorticotrophic hormone
ADCC	Antibody-dependent cell-mediated cytotoxicity
ADP/O ratio	Adenosine diphosphate/oxygen ratio
AIDS	Acquired immune deficiency syndrome
AIHA	American Industrial Hygiene Association
AII	Angiotensin II
ALA	Aminolevulinic acid
ALA-D	Aminolevulinic acid dehydrase
ALA-S	Aminolevulinic acid synthetase
ALA-U	Aminolevulinic acid in urine
APDC	Ammonium pyrrolidine-dithiocarbamate
APHA	American Public Health Association
ASTM	Amercian Society for Testing and Materials
ASV	Anodic stripping voltammetry
ATP	Adenosine triphosphate
B-cells	Bone marrow-derived lymphocytes
Ba	Barium
BAL	British anti-Lewisite (AKA dimercaprol)
BAP	benzo(a)pyrene
BSA	Bovine serum albumin
BUN	Blood serum urea nitrogen
BW	Body weight
C.V.	Coefficient of variation
CaBP	Calcium binding protein
CaEDTA	Calcium ethylenediaminetetraacetate
$CaNa_2EDTA$	Calcium sodium ethylenediaminetetraacetate
CBD	Central business district
Cd	Cadmium
CDC	Centers for Disease Control
CEC	Cation exchange capacity
CEH	Center for Environmental Health
CFR	reference method
CMP	Cytidine monophosphate
CNS	Central nervous system
CO	Carbon monoxide
COHb	Carboxyhemoglobin
CP-U	Urinary coproporphyrin
C_{pah}	plasma clearance of p-aminohippuric acid
Cu	Copper
D.F.	Degrees of freedom
DA	Dopamine
δ-ALA	delta-aminolevulinic acid
DCMU	[3-(3,4-dichlorophenyl)-1,1-dimethylurea
DPP	Differential pulse polarography
DNA	Deoxyribonucleic acid
DTH	Delayed-type hypersensitivity
EEC	European Economic Community
EEG	Electroencephalogram
EMC	Encephalomyocarditis
EP	Erythrocyte protoporphyrin

LIST OF ABBREVIATIONS (continued).

EPA	U.S. Environmental Protection Agency
FA	Fulvic acid
FDA	Food and Drug Administration
Fe	Iron
FEP	Free erythrocyte protoporphyrin
FY	Fiscal year
G.M.	Grand mean
G-6-PD	Glucose-6-phosphate dehydrogenase
GABA	Gamma-aminobutyric acid
GALT	Gut-associated lymphoid tissue
GC	Gas chromatography
GFR	Glomerular filtration rate
HA	Humic acid
Hg	Mercury
hi-vol	High-volume air sampler
HPLC	High-performance liquid chromatography
i.m.	Intramuscular (method of injection)
i.p.	Intraperitoneally (method of injection)
i.v.	Intravenously (method of injection)
IAA	Indol-3-ylacetic acid
IARC	International Agency for Research on Cancer
ICD	International classification of diseases
ICP	Inductively coupled plasma emission spectroscopy
IDMS	Isotope dilution mass spectrometry
IF	Interferon
ILE	Isotopic Lead Experiment (Italy)
IRPC	International Radiological Protection Commission
K	Potassium
LDH-X	Lactate dehydrogenase isoenzyme x
LC$_{50}$	Lethyl concentration (50 percent)
LD$_{50}$	Lethal dose (50 percent)
LH	Luteinizing hormone
LIPO	Laboratory Improvement Program Office
ln	Natural logarithm
LPS	Lipopolysaccharide
LRT	Long range transport
mRNA	Messenger ribonucleic acid
ME	Mercaptoethanol
MEPP	Miniature end-plate potential
MES	Maximal electroshock seizure
MeV	Mega-electron volts
MLC	Mixed lymphocyte culture
MMD	Mass median diameter
MMAD	Mass median aerodynamic diameter
Mn	Manganese
MND	Motor neuron disease
MSV	Moloney sarcoma virus
MTD	Maximum tolerated dose
n	Number of subjects or observations
N/A	Not Available

LIST OF ABBREVIATIONS (continued).

NA	Not Applicable
NAAQS	National ambient air quality standards
NAD	Nicotinamide Adenine Dinucleotide
NADB	National Aerometric Data Bank
NAMS	National Air Monitoring Station
NAS	National Academy of Sciences
NASN	National Air Surveillance Network
NBS	National Bureau of Standards
NE	Norepinephrine
NFAN	National Filter Analysis Network
NFR-82	Nutrition Foundation Report of 1982
NHANES II	National Health Assessment and Nutritional Evaluation Survey II
Ni	Nickel
NTA	Nitrilotriacetonitrile
OSHA	Occupational Safety and Health Administration
P	Phosphorus
p	Significance symbol
PAH	Para-aminohippuric acid
Pb	Lead
PBA	Air lead
$Pb(Ac)_2$	Lead acetate
PbB	concentration of lead in blood
PbBrCl	Lead (II) bromochloride
PBG	Porphobilinogen
PFC	Plaque-forming cells
[illegible]	Measure of acidity
PHA	Phytohemagglutinin
PHZ	Polyacrylamide-hydrous-zirconia
PIXE	Proton-induced X-ray emissions
PMN	Polymorphonuclear leukocytes
PND	Post-natal day
PNS	Peripheral nervous system
P.O.	Per os (orally)
ppm	Parts per million
PRA	Plasma renin activity
PRS	Plasma renin substrate
PWM	Pokeweed mitogen
Py-5-N	Pyrimide-5'-nucleotidase
RBC	Red blood cell; erythrocyte
RBF	Renal blood flow
RCR	Respiratory control ratios/rates
redox	Oxidation-reduction potential
RES	Reticuloendothelial system
RLV	Rauscher leukemia virus
RNA	Ribonucleic acid
S-HT	Serotonin
SA-7	Simian adenovirus
S.C.	Subcutaneously (method of injection)
scm	Standard cubic meter
S.D.	Standard deviation
SDS	Sodium dodecyl sulfate
S.E.M.	Standard error of the mean

LIST OF ABBREVIATIONS (continued).

SES	Socioeconomic status
SGOT	Serum glutamic oxaloacetic transaminase
sIg	Surface immunoglobulin
SLAMS	State and local air monitoring stations
SMR	Standardized mortality ratio
Sr	Strontium
SRBC	Sheep red blood cells
SRMs	Standard reference materials
STEL	Short-term exposure limit
SW voltage	Slow-wave voltage
T-cells	Thymus-derived lymphocytes
t-tests	Tests of significance
TBL	Tri-n-butyl lead
TEA	Tetraethyl-ammonium
TEL	Tetraethyllead
TIBC	Total iron binding capacity
TML	Tetramethyllead
TMLC	Tetramethyllead chloride
TSH	Thyroid-stimulating hormone
TSP	Total suspended particulate
U.K.	United Kingdom
UMP	Uridine monophosphate
USPHS	U.S. Public Health Service
VA	Veterans Administration
V_d	Deposition velocity
VER	Visual evoked response
WHO	World Health Organization
XRF	X-Ray fluorescence
X^2	Chi squared
Zn	Zinc
ZPP	Erythrocyte zinc protoporphyrin

MEASUREMENT ABBREVIATIONS

dl	deciliter
ft	feet
g	gram
g/gal	gram/gallon
g/ha·mo	gram/hectare·month
km/hr	kilometer/hour
l/min	liter/minute
mg/km	milligram/kilometer
$\mu g/m^3$	microgram/cubic meter
mm	millimeter
μm	micrometer
μmol	micromole
ng/cm^2	nanograms/square centimeter
nm	nanometer
nM	nanomole
sec	second
t	tons

GLOSSARY VOLUME II

A horizon of soils - the top layer of soil, immediately below the litter layer; organically rich.

anorexia - loss of appetite.

anthropogenic - generated by the activities of man.

apoplast - extracellular portion of the root cross-section.

Brownian movement - the random movement of microscopic particles.

carnivore - meat-eating organism.

catenation - linkage between atoms of the same chemical element.

cation exchange capacity (CEC) - the ability of a matrix to selectively exchange positively charged ions.

chemical mass balance - the input/output balance of a chemical within a defined system.

coprophilic fungi - fungi which thrive on the biological waste products of other organisms.

detritus - the organic remains of plants and animals.

dictyosome - a portion of the chloroplast structurally similar to a stack of disks.

dry deposition - the transfer of atmospheric particles to surfaces by sedimentation or impaction.

ecosystem - one or more ecological communities linked by a common set of environmental parameters.

electronegativity - a measure of the tendency of an atom to become negatively charged.

enrichment factor - the degree to which the environmental concentration of an element exceeds the expected (natural or crustal) concentration.

galena - natural lead sulfide.

gravimetric - pertaining to a method of chemical analysis in which the concentration of an element in a sample is determined by weight (e.g., a precipitate).

herbivore - plant-eating organism.

humic substances - humic and fulvic acids in soil and surface water.

hydroponically grown plants - plants which are grown with their roots immersed in a nutrient-containing solution instead of soil.

Law of Tolerance - for every environmental factor there is both a minimum and a maximum that can be tolerated by a population of plants or animals.

leaf area index (LAI) - the effective leaf-surface (upfacing) area of a tree as a function of the plane projected area of the tree canopy.

LC_{50} - concentration of an agent at which 50 percent of the exposed population dies.

lithosphere - the portion of the earth's crust subject to interaction with the atmosphere and hydrosphere.

mass median aerodynamic diameter (MMAD) - the aerodynamic diameter (in μm) at which half the mass of particles in an aerosol is associated with values below and half above.

meristematic tissue - growth tissue in plants capable of differentiating into any of several cell types.

microcosm - a small, artificially controlled ecosystem.

mycorrhizal fungi - fungi symbiotic with the root tissue of plants.

NADP - National Atmospheric Deposition Program.

photolysis - decomposition of molecules into simpler units by the application of light.

photosytem I light reaction - the light reaction of photosystem converts light to chemical energy (ATP and reduced NADP). Photosystem I of the light reaction receives excited electrons from photosystem II, increases their energy by the absorption of light, and passes these excited electrons to redox substances that eventually produce reduced NADP.

primary producers - plants and other organisms capable of transforming carbon dioxide and light or chemical energy into organic compounds.

promotional energy - the energy required to move an atom from one valence state to another.

saprotrophs - heterotrophic organisms that feed primarily on dead organic material.

stoichiometry - calculation of the quantities of substances that enter into and are produced by chemical reactions.

stratospheric transfer - in the context of this document, transfer from the troposphere to the stratosphere.

symplast - intracellular portion of the root cross-section.

troposphere - the lowest portion of the atmosphere, bounded on the upper level by the stratosphere.

wet deposition - the transfer of atmospheric particles to surfaces by precipitation, e.g., rain or snow.

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2. INTRODUCTION

According to Section 108 of the Clean Air Act of 1970, as amended in June 1974, a criteria document for a specific pollutant or class of pollutants shall

> . . . accurately reflect the latest scientific knowledge useful in indicating the kind and extent of all identifiable effects on public health or welfare which may be expected from the presence of such pollutant in the ambient air, in varying quantities.

Air quality criteria are of necessity based on presently available scientific data, which in turn reflect the sophistication of the technology used in obtaining those data as well as the magnitude of the experimental efforts expended. Thus air quality criteria for atmospheric pollutants are a scientific expression of current knowledge and uncertainties. Specifically, air quality criteria are expressions of the scientific knowledge of the relationships between various concentrations--averaged over a suitable time period--of pollutants in the same atmosphere and their adverse effects upon public health and the environment. Criteria are issued to help make decisions about the need for control of a pollutant and about the development of air quality standards governing the pollutant. Air quality criteria are descriptive; that is, they describe the effects that have been observed to occur as a result of external exposure at specific levels of a pollutant. In contrast, air quality standards are prescriptive; that is, they prescribe what a political jurisdiction has determined to be the maximum permissible exposure for a given time in a specified geographic area.

In the case of criteria for pollutants that appear in the atmosphere only in the gas phase (and thus remain airborne), the sources, levels, and effects of exposure must be considered only as they affect the human population through inhalation of or external contact with that pollutant. Lead, however, is found in the atmosphere primarily as inorganic particulate, with only a small fraction normally occurring as vapor-phase organic lead. Consequently, inhalation and contact are but two of the routes by which human populations may be exposed to lead. Some particulate lead may remain suspended in the air and enter the human body only by inhalation, but other lead-containing particles will be deposited on vegetation, surface waters, dust, soil, pavements, interior and exterior surfaces of housing--in fact, on any surface in contact with the air. Thus criteria for lead must be developed that will take into account all principal routes of exposure of the human population.

This criteria document is a revision of the previous Air Quality Criteria Document for Lead (EPA-600/8-77-017) published in December, 1977. This revision is mandated by the Clean Air Act (Sections 108 and 109), as amended U.S.C. §§7408 and 7409. The criteria document sets forth what is known about the effects of lead contamination in the environment on human health and welfare. This requires that the relationship between levels of exposure to lead, via all routes and averaged over a suitable time period, and the biological responses to those levels be carefully assessed. Assessment of exposure must take into consideration the temporal and spatial distribution of lead and its various forms in the environment.

This document focuses primarily on lead as found in its various forms in the ambient atmosphere; in order to assess its effects on human health, however, the distribution and biological availability of lead in other environmental media have been considered. The rationale for structuring the document was based primarily on the two major questions of exposure and response. The first portion of the document is devoted to lead in the environment--its physical and chemical properties; the monitoring of lead in various media; sources, emissions, and concentrations of lead; and the transport and transformation of lead within environmental media. The later chapters are devoted to discussion of biological responses and effects on ecosystems and human health.

In order to facilitate printing and distribution of the present materials, this Draft Final version of the revised EPA Air Quality Criteria Document for Lead is being released in the form of four volumes. The first volume (Volume I) contains the executive summary and conclusions chapter (Chapter 1) for the entire document. Volume II (the present volume) contains Chapters 2-8, which include: the introduction for the document (Chapter 2); discussions of the above listed topics concerning lead in the environment (Chapters 3-7); and evaluation of lead effects on ecosystems (Chapter 8). The remaining two volumes contain Chapters 9-13, which deal with the extensive available literature relevant to assessment of health effects associated with lead exposure. In addition to the above materials, there is appended to Chapter 1 an addendum specifically addressing: the complex relationship between blood lead level and blood pressure; and the effects of fetal and pediatric exposures on growth and neurobehavioral development.

An effort has been made to limit the document to a highly critical assessment of the scientific data base through December, 1985. The references cited do not constitute an exhaustive bibliography of all available lead-related literature but they are thought to be sufficient to reflect the current state of knowledge on those issues most relevant to the review of the air quality standard for lead.

The status of control technology for lead is not discussed in this document. For information on the subject, the reader is referred to appropriate control technology documentation published by the Office of Air Quality Planning and Standards (OAQPS), EPA. The subject of adequate margin of safety stipulated in Section 108 of the Clean Air Act also is not explicitly addressed here; this topic will be considered in depth by EPA's Office of Air Quality Planning and Standards in documentation prepared as a part of the process of revising the National Ambient Air Quality Standard for Lead.

3. CHEMICAL AND PHYSICAL PROPERTIES

3.1 INTRODUCTION

Lead is a gray-white metal of silvery luster that, because of its easy isolation and low melting point (327.5°C), was among the first of the metals to be placed in the service of civilization. The Phoenicians traveled as far as Spain and England to mine lead as early as 2000 B.C. The Egyptians also used lead extensively; the British Museum contains a lead figure found in an Egyptian temple which possibly dates from 3000 B.C. The most abundant ore is galena, in which lead is present as the sulfide (PbS); metallic lead is readily smelted from galena. The metal is soft, malleable, and ductile, a poor electrical conductor, and highly impervious to corrosion. This unique combination of physical properties has led to its use in piping and roofing, and in containers for corrosive liquids. By the time of the Roman Empire, it was already in wide use in aqueducts and public water systems, as well as in cooking and storage utensils. Solder, type metal, and various antifriction materials are manufactured from alloys of lead. Metallic lead and lead dioxide are used in storage batteries, and metallic lead is used in cable covering, plumbing and ammunition. Because of its high nuclear cross section, the lead atom can absorb a broad range of radiation, making this element an effective shield around X-ray equipment and nuclear reactors.

This chapter does not attempt to describe all of the properties of lead for each environmental medium. Additional discussions of the chemical properties of lead, as they pertain to specific media such as air and soil, may be found in chapters 6 and 8.

3.2 ELEMENTAL LEAD

In comparison with the most abundant metals in the earth's crust (aluminum and iron), lead is a rare metal; even copper and zinc are more abundant by factors of five and eight, respectively. Lead is, however, more abundant than the other toxic heavy metals; its abundance in the earth's crust has been estimated (Moeller, 1952) to be as high as 160 µg/g, although some other authors (Heslop and Jones, 1976) suggest a lower value of 20 µg/g. Either of these estimates suggests that the abundance of lead is more than 100 times that of cadmium or mercury, two other significant systemic metallic poisons. More important, since lead occurs in highly concentrated ores from which it is readily separated, the availability of lead is far greater than its natural abundance would suggest. The environmental significance of lead is the result both of its utility and of its availability to mankind. Lead ranks fifth among metals in tonnage consumed, after iron, copper, aluminum and zinc; it is, therefore, produced in far larger quantities than any other toxic heavy metal (Dyrssen, 1972). The properties of elemental lead are summarized in Table 3-1.

TABLE 3-1. PROPERTIES OF ELEMENTAL LEAD

Property	Description
Atomic weight	207.19
Atomic number	82
Oxidation states	+2, +4
Density	11.35 g/cm^3 at 20 °C
Melting point	327.5 °C
Boiling point	1740 °C
Covalent radius (tetradehral)	1.44 Å
Ionic radii	1.21 Å (+2), 0.78 Å (+4)
Resistivity	21.9 x 10^{-6} ohm/cm

Natural lead is a mixture of four stable isotopes: ^{204}Pb (~1.5 percent), ^{206}Pb (23.6 percent), ^{207}Pb (22.6 percent), and ^{208}Pb (52.3 percent). There is no radioactive progenitor for ^{204}Pb, but ^{206}Pb, ^{207}Pb, and ^{208}Pb are produced by the radioactive decay of ^{238}U, ^{235}U, and ^{232}Th, respectively. There are four radioactive isotopes of lead that occur as members of these decay series. Of these, only ^{210}Pb is long lived, with a half-life of 22 years. The others are ^{211}Pb (half-life 36.1 min), ^{212}Pb (10.64 hr), and ^{214}Pb (26.8 min). The stable isotopic compositions of naturally occurring lead ores are not identical, but show variations reflecting geological evolution (Russell and Farquhar, 1960). Thus, the observed isotopic ratios depend upon the U/Pb and Th/Pb ratios of the source from which the ore is derived and the age of the ore deposit. The ^{206}Pb/^{204}Pb isotopic ratio, for example, varies from approximately 16.5 to 21 depending on the source (Doe, 1970). The isotopic ratios in average crustal rock reflect the continuing decay of uranium and thorium. The differences between crustal rock and ore bodies, and between major ore bodies in various parts of the world, often permit the identification of the source of lead in the environment.

3.3 GENERAL CHEMISTRY OF LEAD

Lead is the heaviest element in Group IVB of the periodic table; this is the group that also contains carbon, silicon, germanium, and tin. Unlike the chemistry of carbon, however, the inorganic chemistry of lead is dominated by the divalent (+2) oxidation state rather than the tetravalent (+4) oxidation state. This important chemical feature is a direct result of the fact that the strengths of single bonds between the Group IV atoms and other atoms generally decrease as the atomic number of the Group IV atom increases (Cotton and Wilkinson, 1980). Thus, the average energy of a C-H bond is 100 kcal/mole, and it is this factor that stabilizes CH_4 relative to CH_2; for lead, the Pb-H energy is only approximately 50 kcal/mole (Shaw and Allred, 1970), and this is presumably too small to compensate for the Pb(II) → Pb(IV) promotional energy. It is this same feature that explains the marked difference in the tendencies to catenation shown by these elements. Though C-C bonds are present in literally millions of compounds, lead catenation occurs only in organolead compounds. Lead does, however, form compounds like Na_4Pb_9 which contain distinct polyatomic lead clusters (Britton, 1964), and Pb-Pb bonds are found in the cationic cluster $[Pb_6O(OH)_6]^{+4}$ (Olin and Soderquist, 1972).

A listing of the solubilities and physical properties of the more common compounds of lead is given in Appendix 3A (Table 3A-1) (Weast, 1982). As can be discerned from those data, most inorganic lead salts are sparingly soluble (e.g., PbF_2, $PbCl_2$) or virtually insoluble ($PbSO_4$, $PbCrO_4$) in water; the notable exceptions are lead nitrate, $Pb(NO_3)_2$, and lead acetate, $Pb(OCOCH_3)_2$. Inorganic lead (II) salts are, for the most part, relatively high-melting-point solids with correspondingly low vapor pressures at room temperatures. The vapor pressures of the most commonly encountered lead salts are also tabulated in Appendix 3A (Table 3A-2) (Stull, 1947). The transformation of lead salts in the atmosphere is discussed in Chapter 6.

3.4 ORGANOMETALLIC CHEMISTRY OF LEAD

The properties of organolead compounds (i.e., compounds containing bonds between lead and carbon) are entirely different from those of the inorganic compounds of lead; although a few organolead(II) compounds, such as dicyclopentadienyllead, $Pb(C_5H_5)_2$, are known, the organic chemistry of lead is dominated by the tetravalent (+4) oxidation state. An important property of most organolead compounds is that they undergo photolysis when exposed to light (Rufman and Rotenberg, 1980).

Because of their use as antiknock agents in gasoline and other fuels, the most important organolead compounds have been the tetraalkyl compounds tetraethyllead (TEL) and tetramethyllead (TML). As would be expected for such nonpolar compounds, TEL and TML are insoluble in water but soluble in hydrocarbon solvents (e.g., gasoline). These two compounds are manufactured by the reaction of the alkyl chloride with lead-sodium alloy (Shapiro and Frey, 1968):

$$4NaPb + 4C_2H_5Cl \rightarrow (C_2H_5)_4Pb + 3Pb + 4NaCl \qquad (3\text{-}1)$$

The methyl compound, TML, is also manufactured by a Grignard process involving the electrolysis of lead pellets in methylmagnesium chloride (Shapiro and Frey, 1968):

$$2CH_3MgCl + 2CH_3Cl + Pb \rightarrow (CH_3)_4Pb + 2MgCl_2 \qquad (3\text{-}2)$$

A common type of commercial antiknock mixture contains a chemically redistributed mixture of alkyllead compounds. In the presence of Lewis acid catalysts, a mixture of TEL and TML undergoes a redistribution reaction to produce an equilibrium mixture of the five possible tetraalkyllead compounds. For example, an equimolar mixture of TEL and TML produces a product with a composition as shown below:

Component	Mol percent
$(CH_3)_4Pb$	4.6
$(CH_3)_3Pb(C_2H_5)$	24.8
$(CH_3)_2Pb(C_2H_5)_2$	41.2
$(CH_3)Pb(C_2H_5)_3$	24.8
$(C_2H_5)_4Pb$	4.6

These lead compounds are removed from internal combustion engines by a process called lead scavenging, in which they react in the combustion chamber with halogenated hydrocarbon additives (notably ethylene dibromide and ethylene dichloride) to form lead halides, usually bromochlorolead(II). Mobile source emissions are discussed in detail in Section 5.3.3.2.

Several hundred other organolead compounds have been synthesized, and the properties of many of them are reported by Shapiro and Frey (1968). The continuing importance of organolead chemistry is demonstrated by a variety of recent publications investigating the syntheses (Hager and Huber, 1980; Wharf et al., 1980) and structures (Barkigia et al., 1980) of organolead complexes, and by recent patents for lead catalysts (Nishikido et al., 1980).

3.5 FORMATION OF CHELATES AND OTHER COMPLEXES

The bonding in organometallic derivatives of lead is principally covalent rather than ionic because of the small difference in the electronegativities of lead (1.8) and carbon (2.6). As is the case in virtually all metal complexes, however, the bonding is of the donor-acceptor type, in which both electrons in the bonding orbital originate from the carbon atom.

The donor atoms in a metal complex could be almost any basic atom or molecule; the only requirement is that a donor, usually called a ligand, must have a pair of electrons available for bond formation. In general, the metal atom occupies a central position in the complex, as exemplified by the lead atom in tetramethyllead (Figure 3-1a) which is tetrahedrally surrounded by four methyl groups. In these simple organolead compounds, the lead is usually present as Pb(IV), and the complexes are relatively inert. These simple ligands, which bind to metal at only a single site, are called monodentate ligands. Some ligands, however, can bind to the metal atom by more than one donor atom, so as to form a heterocyclic ring structure. Rings of this general type are called chelate rings, and the donor molecules which form them are called polydentate ligands or chelating agents. In the chemistry of lead, chelation normally involves Pb(II), leading to kinetically quite labile (although thermodynamically stable) octahedral complexes. A wide variety of biologically significant chelates with ligands, such as amino acids, peptides, nucleotides and similar macromolecules, are known. The simplest structure of this type occurs with the amino acid glycine, as represented in Figure 3-1b for a 1:2 (metal:ligand) complex. The importance of chelating agents in the present context is their widespread use in the treatment of lead and other metal poisoning.

Metals are often classified according to some combination of their electronegativity, ionic radius and formal charge (Ahrland, 1966, 1968, 1973; Basolo and Pearson, 1967; Nieboer and Richardson, 1980; Pearson, 1963, 1968). These parameters are used to construct empirical classification schemes of relative hardness or softness. In these schemes, "hard" metals form strong bonds with "hard" anions and likewise "soft" metals with "soft" anions. Some metals are borderline, having both soft and hard character. Pb(II), although borderline, demonstrates primarily soft character (Figure 3-2) (Nieboer and Richardson, 1980). The terms Class A and Class B may also be used to refer to hard metals and soft metals, respectively. Since Pb(II) is a relatively soft (or class B) metal ion, it forms strong bonds to soft donor atoms like the sulfur atoms in the cysteine residues of proteins and enzymes; it also coordinates strongly with the imidazole groups of histidine residues and with the carboxyl groups of glutamic and aspartic acid residues. In living systems, therefore, lead atoms bind to these peptide residues in proteins, thereby preventing the proteins from carrying out their functions by changing the tertiary structure of the protein or by blocking the substrate's approach to the active site of the protein. As has been demonstrated in several studies (Jones and Vaughn, 1978; Williams and Turner, 1981; Williams et al., 1982), there is an inverse correlation between the LD_{50} values of metal complexes and the chemical softness parameter (σp) (Pearson and Mawby, 1967). Thus, for both mice and *Drosophila*, soft metal ions like lead(II) have been found to be more toxic than hard metal ions (Williams et al., 1982). This classification of metal ions according to their toxicity has been discussed in detail by Nieboer and Richardson (1980). Lead(II) has a higher softness parameter than either cadmium(II) or mercury(II), so lead(II) compounds would not be expected to be as toxic as their cadmium or mercury analogues.

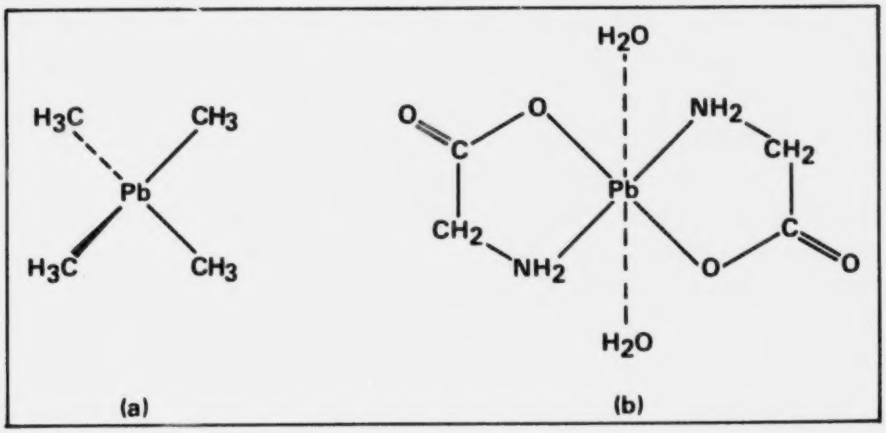

H3C
CH3
Pb
H3C
CH3
(a)
H2O
O
C
O
NH2
CH2
Pb
CH2
NH2
C
O
O
H2O
(b)

Figure 3-1. Metal complexes of lead.

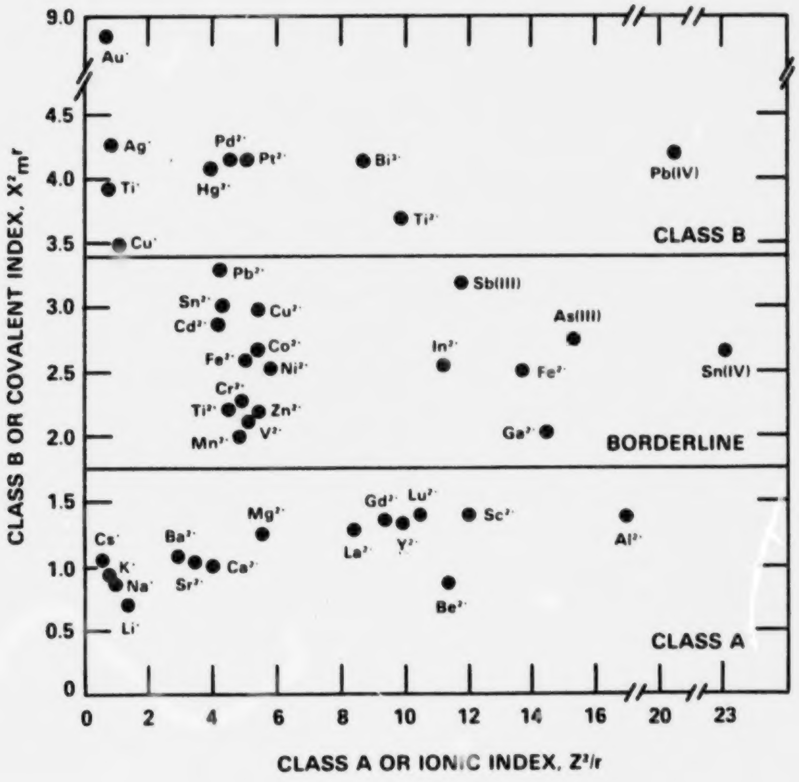
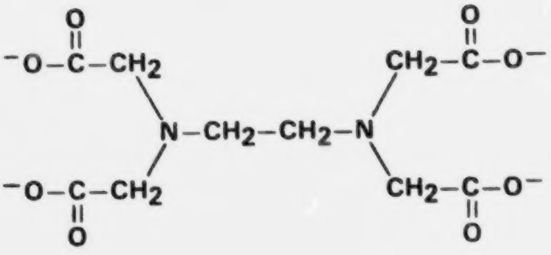
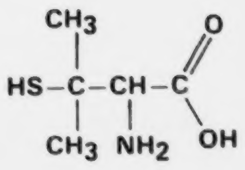


Figure 3-2. Softness parameters of metals.

Source: Nieboer and Richardson (1980).



EDTA



PENICILLAMINE

Figure 3-3. Structure of chelating agents.

The role of the chelating agents is to compete with the peptides for the metal by forming stable chelate complexes that can be transported from the protein and eventually be excreted by the body. For simple thermodynamic reasons (see Appendix 3A), chelate complexes are much more stable than monodentate metal complexes, and it is this enhanced stability that is the basis for their ability to compete favorably with proteins and other ligands for the metal ions. The chelating agents most commonly used for the treatment of lead poisoning are ethylenediaminetetraacetate ions (EDTA), D-penicillamine (Figure 3-3) and their derivatives. EDTA is known to act as a hexadentate ligand toward metals (Lis, 1978; McCandlish et al., 1978). X-ray diffraction studies have demonstrated that D-penicillamine is a tridentate ligand binding through its sulfur, nitrogen and oxygen atoms to cobalt (de Meester and Hodgson, 1977a; Helis et al., 1977), chromium (de Meester and Hodgson, 1977b), cadmium (Freeman et al., 1976), and lead itself (Freeman et al., 1974), but both penicillamine and other cysteine derivatives may act as bidentate ligands (Carty and Taylor, 1977; de Meester and Hodgson, 1977c). Moreover, penicillamine binds to mercury only through its sulfur atoms (Wong et al., 1973; Carty and Taylor, 1976).

It should be noted that both the stoichiometry and structures of metal chelates depend upon pH, and that structures different from those manifest in solution may occur in crystals. It will suffice to state, however, that several ligands can be found that are capable of sufficiently strong chelation with lead present in the body under physiological conditions to permit their use in the effective treatment of lead poisoning.

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APPENDIX 3A

PHYSICAL/CHEMICAL DATA FOR LEAD COMPOUNDS

3A.1 DATA TABLES

TABLE 3A-1. PHYSICAL PROPERTIES OF INORGANIC LEAD COMPOUNDS

					Solubility, g/100 ml		
Compound	Formula	M.W.	S.G.	M.P. (°C)	Cold water	Hot water	Other solvents
Lead	Pb	207.19	11.35	327.5	i	i	sa
Acetate	$Pb(C_2H_3O_2)_2$	325.28	3.25	280	44.3	221^{50}	s glyc
Azide	$Pb(N_3)_2$	291.23	-	expl	0.023	0.09^{70}	-
Bromate	$Pb(BrO_3)_2 \cdot H_2O$	481.02	5.53	d180	1.38	sl s	-
Bromide	$PbBr_2$	367.01	6.66	373	0.8441	4.71^{100}	sa
Carbonate	$PbCO_3$	267.20	6.6	d315	0.00011	d	sa,alk
Carbonate, basic	$2PbCO_3 \cdot Pb(OH)_2$	775.60	6.14	d400	i	i	s HNO_3
Chloride	$PbCl_2$	278.10	5.85	501	0.99	3.34^{100}	i al
Chlorobromide[1]	PbClBr	322.56	-	430	0.66^{19}	1.03^{43}	-
Chromate	$PbCrO_4$	323.18	6.12	844	$6x10^{-6}$	i	sa,alk
Chromate, basic	$PbCrO_4 \cdot PbO$	546.37	6.63	-	i	i	sa,alk
Cyanide	$Pb(CN)_2$	259.23	-	-	sl s	s	s KCN
Fluoride	PbF_2	245.19	8.24	855	0.064	-	s HNO_3
Fluorochloride	PbFCl	261.64	7.05	601	0.037	0.1081	-
Formate	$Pb(CHO_2)_2$	297.23	4.63	d190	1.6	20	i al
Hydride	PbH_2	209.21	-	^{1}d	-	-	-
Hydroxide	$Pb(OH)_2$	241.20	-	d145	0.0155	sl s	sa,alk
Iodate	$Pb(IO_3)_2$	557.00	6.155	d300	0.0012	0.003	s HNO_3
Iodide	PbI_2	461.00	6.16	402	0.063	0.41	s,alk
Nitrate	$Pb(NO_3)_2$	331.20	4.53	d470	37.65	127	s,alk

TABLE 3A-1. (continued)

					Solubility, g/100 ml		
Compound	Formula	M.W.	S.G.	M.P. (°C)	Cold water	Hot water	Other solvents
Nitrate, basic	$Pb(OH)NO_3$	286.20	5.93	d180	19.4	s	sa
Oxalate	PbC_2O_4	295.21	5.28	d300	0.00016	-	sa
Oxide	PbO	223.19	9.53	888	0.0017	-	s,alk
Dioxide	PbO_2	239.19	9.375	d290	i	i	sa
Oxide (red)	Pb_3O_4	685.57	9.1	d500	i	i	sa
Phosphate	$Pb_3(PO_4)_2$	811.51	7	1014	1.4×10^{-5}	i	s,alk
Sulfate	$PbSO_4$	303.25	6.2	1170	0.00425	0.0056	-
Sulfide	PbS	239.25	7.5	1114	8.6×10^{-5}	-	sa
Sulfite	$PbSO_3$	287.25	-	d	i	i	sa
Thiocyanate	$Pb(SCN)_2$	323.35	3.82	d190	0.05	0.2	s,alk

[1] Melting point and solubility data from Corrin and Natusch (1977)

Abbreviations: a - acid; al - alcohol; alk - alkali; d - decomposes;
expl - explodes; glyc - glycol; i - insoluble; s - soluble;
sl s - slightly soluble; M.W. - molecular weight;
S.G. - specific gravity; and M.P. - melting point.

Source: Weast, 1982.

TABLE 3A-2. TEMPERATURE AT WHICH SELECTED LEAD COMPOUNDS REACH DESIGNATED VAPOR PRESSURES

Name	Formula	M.P. (°C)	Vapor Pressure (mm Hg)					
			1 mm	10 mm	40 mm	100 mm	400 mm	760 mm
Lead	Pb	327.4	973°C	1162°C	1309°C	1421°C	1630°C	1744°C
Lead bromide	$PbBr_2$	373	513	610	686	745	856	914
Lead chloride	$PbCl_2$	501	547	648	725	784	893	954
Lead fluoride	PbF_2	855	solid	904	1003	1080	1219	1293
Lead iodide	PbI_2	402	479	571	644	701	807	872
Lead oxide	PbO	890	943	1085	1189	1265	1402	1472
Lead sulfide	PbS	1114	852 (solid)	975 (solid)	1048 (solid)	1108 (solid)	1221	1281

Source: Stull, 1947.

3A.2. THE CHELATE EFFECT

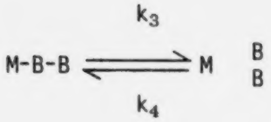
The stability constants of chelated complexes are normally several orders of magnitude higher than those of comparable monodentate complexes; this effect is called the chelate effect, and is very readily explained in terms of kinetic considerations. A comparison of the binding of a single bidentate ligand with that of two molecules of a chemically similar monodentate ligand shows that, for the monodentate case, the process can be represented by the equations:

$$M + B \xrightleftharpoons[k_b]{k_a} M\text{-}B \qquad (3A\text{-}1)$$

$$M\text{-}B + B \xrightleftharpoons[k_d]{k_c} MB_2 \qquad (3A\text{-}2)$$

The related expressions for the bidentate case are:

$$M + B\text{-}B \xrightleftharpoons[k_2]{k_1} M\text{-}B\text{-}B \qquad (3A\text{-}3)$$



$$M\text{-}B\text{-}B \underset{k_4}{\overset{k_3}{\rightleftharpoons}} M\begin{matrix}B\\B\end{matrix} \qquad (3A\text{-}4)$$

The overall equilibrium constants, therefore, are:

$$K_1 = \frac{k_a k_c}{k_b k_d}; \qquad K_2 = \frac{k_1 k_3}{k_2 k_4}$$

For a given metal, M, and two ligands, B and B-B, which are chemically similar, it is established that k_1 and k_a have similar values to each other, as do k_2 and k_b and k_4 and k_d; each of these pairs of terms represents chemically similar processes. The origin of the chelate effect lies in the very large value of k_3 relative to that of k_c. This comes about because k_3 represents a unimolecular process, whereas k_c is a bimolecular rate constant. Consequently, $K_2 >> K_1$.

This concept can, of course, be extended to polydentate ligands; in general, the more extensive the chelation, the more stable the metal complex. Hence, one would anticipate, correctly, that polydentate chelating agents such as penicillamine or EDTA can form extremely stable complexes with metal ions.

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4. SAMPLING AND ANALYTICAL METHODS FOR ENVIRONMENTAL LEAD

4.1 INTRODUCTION

Lead, like all criteria pollutants, has a designated reference method for monitoring and analysis as required in State Implementation Plans for determining compliance with the lead National Ambient Air Quality Standard. The reference method [C.F.R. (1982) 40:§50] uses a high volume sampler (hi-vol) for sample collection and atomic absorption spectrometry for analysis. Inductively coupled plasma emission spectroscopy and X-ray fluorescence are also reference methods for analysis. These and several other analytical procedures are discussed in this chapter. The reference method for sample collection may be revised to require collection of a specific size fraction of atmospheric particles.

Airborne lead originates principally from manmade sources (about 75 - 90 percent comes from automobile exhaust; see Section 5.3.3.1) and is transported through the atmosphere to vegetation, soil, water, and animals. Knowledge of environmental concentrations of lead and the extent of its movement among various media is essential to control lead pollution and assess its effects on human populations.

The collection and analysis of environmental samples for lead require a rigorous quality assurance program [C.F.R. (1982) 40:§58]. It is essential that the investigator recognize all sources of contamination and use every precaution to eliminate them. Potential lead contamination occurs on the surfaces of collection containers and devices, on the hands and clothing of the investigator, in the chemical reagents, in the laboratory atmosphere, and on the labware and tools used to prepare the sample for analysis. General procedures for controlling this contamination of samples in trace metal analysis are described by Zief and Mitchell (1976); specific details are given in Patterson and Settle (1976). In the following discussion of methods for sampling and analysis, it is assumed that all procedures are carried out with precise attention to contamination control.

In the following sections, the specific operation, procedure and instrumentation involved in monitoring and analyzing environmental lead are discussed. Site selection criteria are treated only briefly, due to the lack of verifying data. Much remains to be done in establishing valid criteria for sampler location. The various types of samples and substrates used to collect airborne lead are described. Methods for collecting dry deposition, wet deposition, and aqueous, soil, and vegetation samples are also reviewed along with current sampling methods specific to mobile and stationary sources. Finally, advantages and limitations of techniques for sample preparation and analysis are discussed.

4.2 SAMPLING

The purpose of sampling is to determine the nature and concentration of lead in the environment. Sampling strategy is dictated by research needs. This strategy encompasses site selection, choice of instrument used to obtain representative samples, and choice of method used to preserve sample integrity. In the United States, sampling stations for air pollutants have been operated since the early 1950's. These early stations were a part of the National Air Surveillance Network (NASN), which has now become the National Filter Analysis Network (NFAN). Two other types of networks have been established to meet specific data requirements. State and Local Air Monitoring Stations (SLAMS) provide data from specific areas where pollutant concentrations and population densities are the greatest and where monitoring of compliance to standards is critical. The National Air Monitoring Station (NAMS) network is designed to serve national monitoring needs, including assessment of national ambient trends. SLAMS and NAMS stations are maintained by state and local agencies and the air samples are analyzed in their laboratories. Stations in the NFAN network are maintained by state and local agencies, but the samples are analyzed by laboratories in the U.S. Environmental Protection Agency, where quality control procedures are rigorously maintained.

Data from all three networks are combined into one data base, the National Aerometric Data Bank (NADB). These data may be individual chemical analyses of a 24-hour sampling period arithmetically averaged over a calendar period, or chemical composites of several filter samples used to determine a quarterly composite. Data are occasionally not available because they do not conform to strict statistical requirements. A summary of the data from the NADB appears in Section 7.2.1.

4.2.1 Regulatory Siting Criteria for Ambient Aerosol Samplers

In September of 1981, EPA promulgated regulations establishing ambient air monitoring and data reporting requirements for lead [C.F.R. (1982) 40:§58] comparable to those already established in May of 1979 for the other criteria pollutants. Whereas sampling for lead is accomplished when sampling for total suspended particulates (TSP), the designs of lead and TSP monitoring stations must be complementary to insure compliance with the NAMS criteria for each pollutant, as presented in Table 4-1, Table 4-2, and Figure 4-1.

In general, the criteria with respect to monitoring stations designate that there must be at least two SLAMS sites for lead in any area which has a population greater than 500,000 and/or any area where lead concentration currently exceeds the ambient lead standard (1.5 $\mu g/m^3$) or has exceeded it since January 1, 1974. In such areas, the SLAMS sites designated as part of the NAMS network must include a microscale or middlescale site located near a major roadway [≥30,000 average daily traffic (ADT)], as well as a neighborhood scale site located in a highly populated residential sector with high traffic density (≥30,000 ADT).

TABLE 4-1. DESIGN OF NATIONAL AIR MONITORING STATIONS

	Conditions	Spatial scale	Minimum number of stations required	Traffic density	Required Siting of Station	
					Meters from edge of roadway	Meters above ground level
Category A						
TSP	High traffic and population density	Neighborhood	see Table 4-2	≧30000	see Figure 4-1	
Pb	Major roadway	Microscale	One	≧30000	5-15	2-7
	Major roadway	Middlescale	One	≦10000	15-50	2-15
				20000	15-75	2-15
				≧40000	15-100	2-15
Category B						
Pb	High traffic and population density	Neighborhood	One	≦10000	> 50	2-15
				20000	> 75	2-15
				≧40000	>100	2-15

Source: C.F.R. (1982) 40:§58 App E.

TABLE 4-2. TSP NAMS CRITERIA

Population Category	Approximate number of stations per area Concentration High[1]	 Medium[2]	 Low[3]
High -- >500,000	6-8	4-6	0-2
Medium -- 100-500,000	4-6	2-4	0-2
Low -- 50-100,000	2-4	1-2	0

[1] When TSP concentration exceeds by 20% Primary Ambient Air Standard of 75 μg/m^3 annual geometric mean.

[2] TSP concentration > Secondary Ambient Air Standard of 60 μg/m^3 annual geometric mean.

[3] TSP concentration < Secondary Ambient Air Standard.

Source: C.F.R. (1982) 40:§58 App D.

With respect to the siting of monitors for lead and other criteria pollutants, there are standards for elevation of the monitors above ground level, setback from roadways, and setback from obstacles. A summary of the specific siting requirements for lead is presented in Table 4-1 and summarized below:

- Samples must be placed between 2 and 15 meters from the ground and greater than 20 meters from trees.
- Spacing of samplers from roads should vary with traffic volume; a range of 5 to 100 meters from the roadway is suggested.
- Distance from samplers to obstacles must be at least twice the height the obstacle protrudes above the sampler.
- There must be a 270° arc of unrestricted air flow around the monitor to include the prevailing wind direction that provides the maximum pollutant concentration to the monitor.
- No furnaces or incineration flues should be in close proximity to the monitor.

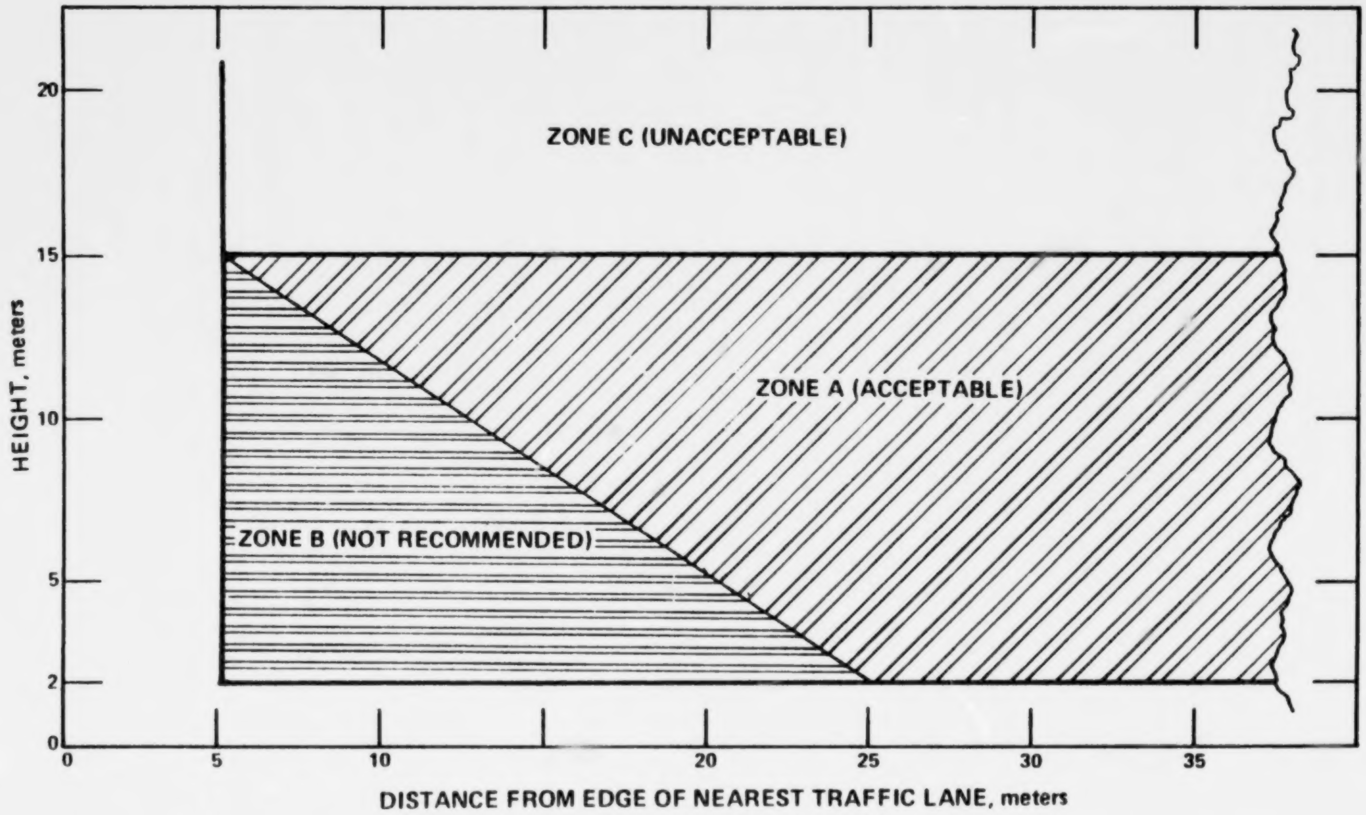

Figure 4-1. Acceptable zone for siting TSP monitors where the average daily traffic exceeds 3000 vehicles/day.

Zone A: Recommended for neighborhood, urban, regional and most middle spatial scales. All NAMS are in this zone.
Zone B: If SLAMS are placed in Zone B they have middle scale of representativeness.

Source: C.F.R. (1982) 40: § 58

To clarify the relationship between monitoring objectives and the actual siting of a monitor, the concept of a spatial scale of representativeness was developed. The spatial scales are described in terms of the physical dimensions of the air space surrounding the monitor throughout which pollutant concentrations are fairly similar. Table 4-3 describes the scales of representativeness while Table 4-4 relates monitoring objectives to the appropriate spatial scale [C.F.R. (1982) 40:§58].

The time scale may also be an important factor. A study by Lynam (1972) illustrates the effect of setback distance on short-term (15-minute) measurements of lead concentrations directly downwind from the source. They found sharp reductions in lead concentration with increasing distance from the roadway. A similar study by PEDCo Environmental, Inc. (1981) did not show the same pronounced reduction when the data were averaged over monthly or quarterly time periods. The apparent reason for this effect is that windspeed and direction are not consistent. Therefore, siting criteria must include sampling times sufficiently long to include average windspeed and direction, or a sufficient number of samples must be collected over short sampling periods to provide an average value consistent with a 24-hour exposure.

4.2.2 Ambient Sampling for Particulate and Gaseous Lead

Airborne lead is primarily inorganic particulate matter (PM) but may occur in the form of organic gases. Devices used for collecting samples of ambient atmospheric lead include the standard hi-vol and a variety of other collectors employing filters, impactors, impingers, or scrubbers, either separately or in combination. Some samplers measure total particulate matter gravimetrically; thus the lead data are usually expressed in μg/g PM or μg/m^3 air. Other samplers do not measure PM gravimetrically; therefore, the lead data can only be expressed as μg/m^3. Some samplers measure lead deposition expressed in μg/cm^2. Some instruments separate particles by size. As a general rule, particles smaller than 2.5 μm are defined as fine, and those larger than 2.5 μm are defined as coarse.

In a typical sampler, the ambient air is drawn down into the inlet and deposited on the collection surface after one or more stages of particle size separation. Inlet effectiveness, internal wall losses, and retention efficiency of the collection surface may bias the collected sample by selectively excluding particles of certain sizes.

4.2.2.1 High Volume Sampler (hi-vol). The present SLAMS and NAMS employ the standard hi-vol sampler (Robson and Foster, 1962; Silverman and Viles, 1948; U.S. Environmental Protection Agency, 1971) as part of their sampling networks. As a Federal Reference Method Sampler, the hi-vol operates with a specific flow rate range of 1.13 - 1.70 m^3/min, drawing air through a

TABLE 4-3. DESCRIPTION OF SPATIAL SCALES OF REPRESENTATIVENESS

Microscale	Defines ambient concentrations in air volumes associated with areas ranging from several to 100 m^2 in size.
Middle scale	Defines concentrations in areas from 100 to 500 m^2 (area up to several city blocks).
Neighborhood scale	Defines concentrations in an extended area of uniform land use, within a city, from 0.5 to 4.0 km^2 in size.
Urban scale	Defines citywide concentrations, areas from 4-50 km^2 in size. Usually requires more than one site.
Regional scale	Defines concentrations in a rural area with homogeneous geography. Range of tens to hundreds of km^2.
National and global scales	Defines concentrations characterizing the U.S. and the globe as a whole.
Personal	Defines air proximate to human respiration, usually sampled with a portable pump.

Source: C.F.R. (1982) 40:§58 App. D; personal scale added in this report.

TABLE 4-4. RELATIONSHIP BETWEEN MONITORING OBJECTIVES AND APPROPRIATE SPATIAL SCALES

Monitoring objective	Appropriate spatial scale for siting air monitors
Highest concentration	Micro, Middle, Neighborhood (sometimes Urban).
Population	Neighborhood, Urban
Source impact	Micro, Middle, Neighborhood
General (background)	Neighborhood, Regional

Source: C.F.R. (1982) 40:§58 App. D.

200 x 250 mm glass fiber filter. At these flow rates, 1600 - 2500 m^3 of air per day are sampled. Many hi-vol systems are presently equipped with mass flow sensors to control the total flow rate through the filter.

The present hi-vol approach has been shown during performance characterization tests to have a number of deficiencies. Wind tunnel testing by Wedding et al. (1977) has shown that the collection characteristics of hi-vol samples are strongly affected by particle size, wind speed and direction, and inlet size. However, since most lead particles have been shown to have a mass median aerodynamic diameter (MMAD) in the range of 0.25 - 1.4 μm (Lee and Goranson, 1972), the hi-vol sampler should present reasonably good estimates of ambient lead concentrations. For particles larger than 5 μm, the hi-vol system is unlikely to collect representative samples (McFarland et al., 1979; Wedding et al., 1977).

4.2.2.2 Dichotomous Sampler. The dichotomous sampler collects two particle size fractions, typically 0 - 2.5 μm and 2.5 μm to the upper cutoff of the inlet employed (normally 10 μm). The impetus for the dichotomy of collection, which approximately separates the fine and coarse particles, was provided by Whitby et al. (1972) to assist in the identification of particle sources. A 2.5 μm cutpoint for the separator was also recommended by Miller et al. (1979) because it satisfied the requirements of health researchers interested in respirable particles, provided adequate separation between two naturally occurring peaks in the size distribution, and was mechanically practical. Because the fine and coarse fractions collected in most locations tend to be acidic and basic, respectively, this separation also minimizes potential particle interaction after collection.

The particle separation principle used by this sampler was described by Hounam and Sherwood (1965) and Conner (1966). The version now in use by EPA was developed by Loo et al. (1979). The separation principle involves acceleration of the particles through a nozzle. Ninety percent of the flowstream is diverted to a small particle collector, while the larger particles continue by inertia toward the large particle collection surface. The inertial virtual impactor design causes 10 percent of the fine particles to be collected with the coarse particle fraction. Therefore, the mass of fine and coarse particles must be adjusted to allow for their cross contamination. This mass correction procedure has been described by Dzubay et al. (1982).

Teflon® membrane filters with pore sizes as large as 2.0 μm can be used in the dichotomous sampler (Dzubay et al, 1982; Stevens et al., 1980) and have been shown to have essentially 100 percent collection efficiency for particles with an aerodynamic diameter as small as 0.03 μm (Liu et al., 1976; see Section 4.2.5). Because the sampler operates at a flowrate of 1 m^3/hr (167 l/min) and collects sub-milligram quantities of particles, a microbalance with a 1 μg resolution is recommended for filter weighing (Shaw, 1980). Removal of the fine particles via this fractionation technique may result in some of the collected coarse particles falling off the filter if care is not taken during filter handling and shipping. However, Dzubay and Barbour (1983) have developed a filter coating procedure which eliminates particle loss during transport. A study by Wedding et al. (1980) has shown that the Sierra® inlet to the dichotomous sampler was sensitive to windspeed. The 50 percent cutpoint (D_{50}) was found to vary from 10 to 22 μm over the windspeed range of 0 to 15 km/hr.

Automated versions of the sampler allow timely and unattended changes of the sampler filters. Depending on atmospheric concentrations, short-term samples of as little as 4 hours can provide diurnal pattern information. The mass collected during such short sample periods, however, is extremely small and highly variable results may be expected.

4.2.2.3 Impactor Samplers. Impactors provide a means of dividing an ambient particle sample into subfractions of specific particle size for possible use in determining size distribution. A jet of air is directed toward a collection surface, which is often coated with an adhesive or grease to reduce particle bounce. Large, high-inertia particles are unable to turn with the airstream; consequently, they hit the collection surface. Smaller particles follow the airstream and are directed toward the next impactor stage or to the filter. Use of multiple stages, each with a different particle size cutpoint, provides collection of particles in several size ranges.

For determining particle mass, removable impaction surfaces may be weighed before and after exposure. The particles collected may be removed and analyzed for individual elements. The selection and preparation of these impaction surfaces have significant effects on the impactor performance. Improperly coated or overloaded surfaces can cause particle bounce to lower stages resulting in substantial cutpoint shifts (Dzubay et al., 1976). Additionally, coatings may cause contamination of the sample. Marple and Willeke (1976) showed the effect of various impactor substrates on the sharpness of the stage cutpoint. Glass fiber substrates can also cause particle bounce or particle interception (Dzubay et al., 1976) and are subject to the formation of artifacts, due to reactive gases interacting with the glass fiber, similar to those on hi-vol sampler filters (Stevens et al., 1978).

Cascade impactors typically have 2 to 10 stages, and flowrates for commercial low-volume versions range from about 0.01 to 0.10 m^3/min. Lee and Goranson (1972) modified a commercially available 0.03 m^3/min low-volume impactor and operated it at 0.14 m^3/min to obtain larger mass collections on each stage. Cascade impactors have also been designed to mount on a hi-vol sampler and operate at flowrates as high as 0.6 - 1.1 m^3/min.

Particle size cutpoints for each stage depend primarily on sampler geometry and flowrate. The smallest particle size cutpoint routinely used is approximately 0.3 μm, although special low-pressure impactors such as that described by Hering et al. (1978) are available with cutpoints as small as 0.05 μm. However, due to the low pressure, volatile organics and nitrates are lost during sampling. A membrane filter is typically used after the last stage to collect the remaining small particles.

4.2.2.4 Dry Deposition Sampling. Dry deposition may be measured directly with surrogate or natural surfaces, or indirectly using micrometeorological techniques. The earliest surrogate surfaces were dustfall buckets placed upright and exposed for several days. The Health and Safety Laboratory (HASL) wet-dry collector is a modification which permits one of a pair of buckets to remain covered except during rainfall. These buckets do not collect a representative sample of particles in the small size range where lead is found because the rim perturbs the natural turbulent flow of the main airstream (Hicks et al., 1980). They are widely used for other pollutants, especially those found primarily on large particles, in the National Atmospheric Deposition Program.

Other surrogate surface devices with smaller rims or no rims have been developed recently (Elias et al., 1976; Lindberg et al., 1979; Peirson et al., 1973). Peirson et al. (1973) used horizontal sheets of filter paper exposed for several days with protection from rainfall. Elias et al. (1976) used Teflon® disks held rigid with a 1 cm Teflon® ring. Lindberg et al. (1979) used petri dishes suspended in a forest canopy. In all of these studies, the calculated deposition velocity (see Section 6.3.1) was within the range expected for small aerosol particles.

A few studies have measured direct deposition on vegetation surfaces using chemical washing techniques to remove surface particles. These determinations are generally 4 - 10 times lower than comparable surrogate surface measurements (Elias et al., 1976; Lindberg et al., 1979), but the reason for this difference could be that natural surfaces represent net accumulation rather than total deposition. Lead removed by rain, dripping dew, or other processes such as foliar uptake would result in an apparently lower deposition rate. In the Lindberg et al. (1979) study, leaves were collected during rainless periods and could not have been influenced by rain washoff. Removal by dew or intercepted fog dripping from the leaves could not be ruled out, but the explanation given by a subsequent report (Lindberg and Harriss, 1981) was that some dry deposition was absorbed by the foliage, that is, foliar uptake was occurring.

There are several micrometeorological techniques that have been used to measure particle deposition. They overcome a deficiency of surrogate surfaces, the lack of correlation between the natural and artificial surfaces, but micrometeorological techniques require expensive equipment and skilled operators. They measure instantaneous or short-term deposition only, and this deposition is inferred to be to a plane-projected surface area only, not necessarily to vegetation surfaces.

Of the five micrometeorological techniques commonly used to measure particle deposition, only two have been used to measure lead particle deposition. Everett et al. (1979) used the profile gradient technique by which lead concentrations are measured at two or more levels within 10 m above the surface. Parallel meteorological data are used to calculate the net flux downward. Droppo (1980) used eddy correlation, which measures fluctuations in the vertical wind component with adjacent measurements of lead concentrations. The calculated differences of each can be used to determine the turbulent flux. These two micrometeorological techniques and the three not yet used for lead, modified Bowen, variance, and eddy accumulation, are described in detail in Hicks et al. (1980).

4.2.2.5 Gas Collection. When sampling ambient lead with systems employing filters, it is likely that vapor-phase organolead compounds will pass through the filter media. The use of bubblers downstream of the filter containing a suitable reagent or absorber for collection of these compounds has been shown to be effective (Purdue et al., 1973). Organolead may be collected on iodine crystals, adsorbed on activated charcoal, or absorbed in an iodine monochloride solution (Skogerboe et al., 1977b).

In one experiment, Purdue et al. (1973) operated two bubblers in series containing iodine monochloride solution. One hundred percent of the lead was recovered in the first bubbler. It should be noted, however, that the analytical detection sensitivity was poor. In general, use of bubblers limits the sample volume (and consequently the sample collection period) due to losses by evaporation and/or bubble carryover. Birch et al. (1980) addressed this problem by increasing the volume of iodine monochloride solution and modifying the inlet impinger to reduce foaming. These authors reported a 97-99 percent collection efficiency of 100 to 500 ng Pb in the first of two bubblers in series. Under ambient sample conditions, this procedure can be used to collect a 24-48 hour sample, provided precautions are taken to retard the decomposition of iodine monochloride by avoiding exposure to light. The sensitivity was 0.25 ng Pb/m^3 for a 48-hour sample.

These procedures do not identify the specific organolead compound collected. Blaszkewicz and Neidhart (1983) have described a technique for the quantitative identification of four organolead compounds: tetramethyl lead, tetraethyl lead, trimethyl lead, and triethyl lead. This technique requires a minimum sample size of 70 ng Pb, which is probably higher than ambient under most collection conditions for a 24- to 48-hour sampling period.

4.2.3 Source Sampling

Sources of atmospheric lead include automobiles, smelters, coal-burning facilities, waste oil combustion, battery manufacturing plants, chemical processing plants, facilities for scrap processing, and welding and soldering operations (see Section 5.3.3). A potentially important secondary source is fugitive dust from mining operations and from soils contaminated with automotive emissions (Olson and Skogerboe, 1975). Chapter 5 contains a complete discussion of sources of lead emissions. The following sections discuss the sampling near potential stationary and mobile sources. Neither indoor nor personal monitoring for lead is performed routinely for ambient situations, although Roy (1977) and Tosteson et al. (1982) discuss the techniques used for personal sampling under special circumstances (see Section 7.2.1.3.3).

4.2.3.1 Stationary Sources. Sampling of stationary sources for lead requires the use of a sequence of samplers at the source of the effluent stream. Since lead in stack emissions may be present in a variety of physical and chemical forms, source sampling trains must be designed to trap and retain both gaseous and particulate lead. A sampling probe is inserted directly in the stack or exhaust stream. In the tentative ASTM method for sampling for atmospheric lead, air is pulled through a 0.45 µm membrane filter and an activated carbon adsorption tube (American Society for Testing and Materials, 1975a).

4.2.3.2 Mobile Sources. Three principal procedures have been used to obtain samples of auto exhaust aerosols for subsequent analysis for lead compounds: a horizontal dilution tunnel, plastic sample collection bags, and a low residence time proportional sampler. In each procedure, samples are air-diluted to simulate roadside exposure conditions. In the most commonly used procedure, a large horizontal air dilution tube segregates fine combustion-derived particles from larger lead particles ablated from combustion chamber and exhaust deposits. In one example of this procedure (Habibi, 1970), hot exhaust is ducted into a 56-cm diameter, 12-m long, air dilution tunnel and mixed with filtered ambient air in a 10-cm diameter mixing baffle in a concurrent flow arrangement. Total exhaust and dilution airflow rate is 28 - 36 m^3/min, which produces a residence time of approximately 5 sec in the tunnel. At the downstream end of the tunnel, samples of the aerosol are obtained by means of isokinetic probes using filters or cascade impactors.

In recent years, various configurations of the horizontal air dilution tunnel have been developed. Several dilution tunnels have been made of polyvinyl chloride with a diameter of 46 cm, but these are subject to wall losses due to charge effects (Gentel et al., 1973; Moran et al., 1972; Trayser et al., 1975). Such tunnels of varying lengths have been limited by exhaust temperatures to total flows above approximately 11 m^3/min. Similar tunnels have a centrifugal fan located upstream, rather than a positive displacement pump located downstream (Trayser et al., 1975). This geometry produces a slight positive pressure in the tunnel and expedites transfer of the aerosol to holding chambers for studies of aerosol growth. However, turbulence from the fan may affect the sampling efficiency. Since the total exhaust plus dilution airflow is not held constant in this system, potential errors can be reduced by maintaining a very high dilution air/exhaust flow ratio (Trayser et al., 1975).

There have also been a number of studies using total filtration of the exhaust stream to arrive at material balances for lead with rather low back-pressure metal filters in an air distribution tunnel (Habibi, 1973; Hirschler et al., 1957; Hirschler and Gilbert, 1964; Sampson and Springer, 1973). The cylindrical filtration unit used in these studies is better than 99 percent efficient in retaining lead particles (Habibi, 1973). Supporting data for lead balances generally confirm this conclusion (Kunz et al., 1975).

In the bag technique, auto emissions produced during simulated driving cycles are air-diluted and collected in a large plastic bag. The aerosol sample is passed through a filtration or impaction sampler prior to lead analysis (Ter Haar et al., 1972). This technique may result in errors of aerosol size analysis because of condensation of low vapor pressure organic substances onto the lead particles.

To minimize condensation problems, a third technique, a low residence time proportional sampling system, has been used. It is based on proportional sampling of raw exhaust, again diluted with ambient air followed by filtration or impaction (Ganley and Springer, 1974; Sampson and Springer, 1973). Since the sample flow must be a constant proportion of the total exhaust flow, this technique may be limited by the response time of the equipment to operating cycle phases that cause relatively small transients in the exhaust flow rate.

4.2.4 Sampling for Lead in Water, Soil, Plants, and Food

Other primary environmental media that may contain airborne lead include precipitation, surface water, soil, vegetation, and foodstuffs. The sampling plans and the sampling methodologies used in dealing with these media depend on the purpose of the experiments, the types of measurements to be carried out, and the analytical technique to be used. General approaches are given below in lieu of specific procedures associated with the numerous possible special situations.

4.2.4.1 Precipitation. Methods developed and used at the Oak Ridge National Laboratory for precipitation collection and analysis for lead are described in Lindberg et al. (1979), Lindberg (1982), and Lindberg and Turner (1983). The investigation should be aware that dry deposition occurs continuously, that lead at the start of a rain event is higher in concentration than at the end, and that rain striking the canopy of a forest may rinse dry deposition particles from the leaf surfaces. Rain collection systems should be designed to collect precipitation on an event basis and to collect sequential samples during the event. They should be tightly sealed from the atmosphere before and after sampling to prevent contamination from dry deposition, falling leaves, and flying insects. Samples for total lead analysis should be acidified to pH less than 2 with nitric acid and refrigerated immediately after sampling. Samples to be separated for particulate and dissolved lead analysis should be filtered prior to acidification. All collection and storage surfaces should be thoroughly cleaned and free of contamination.

Two automated rain-collecting systems have been in use for some time. The Sangamo Precipitation Collector, Type A, collects rain in a single bucket exposed at the beginning of the rain event (Samant and Vaidya, 1982). These authors reported no leaching of lead from the bucket into a solution of 0.3N HNO_3. A second sampler, described by Coscio et al. (1982), also remains covered between rain events; it can collect a sequence of eight samples during the period of rain and may be fitted with a refrigeration unit for sample cooling. No reports of lead analyses were given. Because neither system is widely used for lead sampling, their monitoring effectiveness has not been thoroughly evaluated.

4.2.4.2 Surface Water. Atmospheric lead may be dissolved in water as hydrated ions, chemical complexes, and soluble compounds, or it may be associated with suspended matter. Because the physicochemical form often influences environmental effects, there is a need to differentiate among the various chemical forms of lead. Complete differentiation among all such forms is a complex task that has not yet been fully accomplished. The most commonly used approach is to distinguish between dissolved and suspended forms of lead. All lead passing through a 0.45 µm membrane filter is operationally defined as dissolved, while that retained on the filter is defined as suspended (Kopp and McKee, 1983). Figura and McDuffie (1979, 1980) broadened this scheme to encompass four categories of metal lability that are presumably more representative of uptake by biological systems. These categories are: very labile, moderately labile, slowly labile, and inert. Distinctions between categories are made experimentally by column ion exchange, batch ion exchange, and anodic stripping voltammetry. The key point is the kinetics of the experimental process. If the metal complex can be made to dissociate within milliseconds (anodic stripping voltammetry), then it is considered very labile. Assuming that biological systems take up metals in the free ion state rather than as metal complexes, this scheme can provide important information on the bioavailability of lead in natural waters. Cox et al. (1984) provide evidence that Donnan dialysis, which uses an ion exchange membrane rather than a resin column, may provide a better estimate of lability for lead in natural waters than Chelex-100.

When sampling water bodies, flow dynamics should be considered in the context of the purpose for which the sample is collected. Water at the convergence point of two flowing streams, for example, may not be well mixed for several hundred meters. Similarly, the heavy metal concentrations above and below the thermocline of a lake may be very different. Thus, several samples should be selected in order to define the degree of horizontal or vertical variation. The final sampling plan should be based on the results of pilot studies. In cases where the average concentration is of primary concern, samples can be collected at several points and then mixed to obtain a composite.

Containers used for sample collection and storage should be fabricated from essentially lead-free plastic or glass, e.g., conventional polyethylene, Teflon®, or quartz. These containers must be leached with hot acid for several days to ensure minimum lead contamination (Patterson and Settle, 1976). If only the total lead is to be determined, the sample may be collected without filtration in the field. Nitric acid should be added immediately to reduce the pH to less than 2; the acid will normally dissolve the suspended lead. Otherwise, it is recommended that the sample be filtered upon collection to separate the suspended and dissolved lead and the latter preserved by acid addition as above (U.S. Environmental Protection Agency, 1978). It is also recommended that water samples be stored at 4°C until analysis to avoid further leaching from the container wall (Fishman and Erdmann, 1973; Kopp and Kroner, 1967; Lovering, 1976; National Academy of Sciences, 1972; U.S. Environmental Protection Agency, 1978).

4.2.4.3 Soils. The distance from emission sources and depth gradients associated with lead in soil must be considered in designing the sampling plan. Vegetation, litter, and large objects such as stones should not be included in the sample, depth samples should be collected at 2 cm intervals to preserve vertical integrity, and the samples should be air dried and stored in sealed containers until analyzed. Brown and Black (1983) have addressed the problem of quality assurance and quality control in the collection and analysis of soil samples. A twelve-step procedural protocol and a three-step data validation process were recommended to obtain the most accurate results, and some suggestions were made for handling data bias, precision and uncertainty. Eastwood and Jackson (1984) reported the results of an interlaboratory study that showed greater variations between laboratories than within a laboratory, especially when different analytical procedures are followed.

The chemical similarities between lead complexes in natural waters and in the water associated with soil are not clearly established in the literature. In the more concentrated medium of soil moisture, the lability of lead may change, favoring higher percentages of inert or slowly labile lead (see Section 6.5.1). Although there are many procedures for the analysis of bulk soil samples and for extruding metals from soils in a manner that simulates plant uptake, there are few reports on the collection and analysis of soil moisture at the site of root uptake. The techniques developed by Hinkley and Patterson (1973) for sampling the film of moisture surrounding soil particles have been used by Elias et al. (1976, 1978, 1982) and Elias and Patterson (1980) for the analysis of lead in small volumes of moisture extracted from soil particles in the root zone.

4.2.4.4 Vegetation. Because most soil lead is in forms unavailable to plants, and because lead is not easily transported by plants, roots typically contain very little lead and shoots even less (Zimdahl, 1976; Zimdahl and Koeppe, 1977). Before analysis, a decision must be made as to whether or not the plant material should be washed to remove surface contamination from dry deposition and soil particles. If the plants are sampled for total lead content (e.g., if they represent animal food sources), they cannot be washed. If the effect of lead on internal plant processes is being studied, the plant samples should be washed. In either case, the decision must be made at the time of sampling, as washing cannot be effective after the plant materials have dried. Fresh plant samples cannot be stored for any length of time in a tightly closed container before washing because molds and enzymatic action may affect the distribution of lead on and in the plant tissues. Freshly picked leaves stored in sealed polyethylene bags at room temperature generally begin to decompose in a few days. Storage time may be increased to approximately 2 weeks by refrigeration. Samples that are to be stored for extended periods of time should be oven dried to arrest enzymatic reactions and render the plant tissue amenable to grinding. Storage in sealed containers is required after grinding. For analysis of surface lead, fresh, intact plant parts are agitated in dilute nitric acid or EDTA solutions for a few seconds.

4.2.4.5 Foodstuffs. Analyses for lead in food have been included in the Food and Drug Administration's Total Diet Study since 1972. Initially, this survey involved sampling of foods representing the average diet of a 15 to 20 year-old male, i.e., the individual who on a statistical basis eats the greatest quantity of food (Kolbye et al., 1974). Various food items from the several food classes were purchased in retail stores in various cities across the nation. The foods were cooked or otherwise prepared as they would be in the kitchen, then composited into 12 food classes and analyzed chemically. Other FDA sampling programs are required for different investigative purposes, e.g., enforcement of regulations. For those foods where lead may be deposited on the edible portion, typical kitchen washing procedures are used. This survey procedure has been replaced by one involving separate analyses of 234 individual foods and covering 8 age-sex groups (Pennington, 1983). It is this revised sampling and analytical format that is the basis for food exposure estimates in Section 7.3.1.2.

4.2.5 Filter Selection and Sample Preparation

In sampling for airborne lead, air is drawn through filter materials such as glass fiber, cellulose acetate, or porous plastic (Skogerboe et al., 1977b, Stern, 1976). These materials often include contaminant lead that can interfere with the subsequent analysis (Gandrud and Lazrus, 1972; Kometani et al. 1972; Luke et al., 1972; Seeley and Skogerboe, 1974). If a large mass of particulate matter is collected, then the effects of these trace contaminants may be negligible (Witz and MacPhee, 1976). Procedures for cleaning filters to reduce the lead blank rely on washing with acids or complexing agents (Gandrud and Lazrus, 1972). The type of filter and the analytical method to be used often determines the washing technique.

In some methods, e.g., X-ray fluorescence, analysis can be performed directly on the filter if the filter material is suitable (Dzubay and Stevens, 1975). Skogerboe (1974) provided a general review of filter materials.

The main advantages of glass fiber filters are low pressure drop and high particle collection efficiency at high flow rates. The main disadvantage is variable lead blank, which makes their use inadvisable in many cases (Kometani et al., 1972; Luke at al., 1972). This has placed a high priority on the standardization of a suitable filter for hi-vol samples (Witz and MacPhee, 1976). Other investigations have indicated, however, that glass fiber filters are now available that do not present a lead interference problem (Scott et al., 1976b). Teflon® filters have been used since 1975 by Dzubay et al. (1982) and Stevens et al. (1978), who have shown these filters to have very low lead blanks (<2 ng/cm^2). The collection efficiencies of filters, and also of impactors, have been shown to be dominant factors in the quality of the derived data (Skogerboe et al., 1977a).

Sample preparation usually involves conversion to a solution through wet ashing of solids with acids or through dry ashing in a furnace followed by acid treatment. Either approach works effectively if used properly (Kometani et al., 1972; Skogerboe et al., 1977b). In one investigation of porous plastic Nuclepore® filters, some lead blanks were too high to allow measurements of ambient air lead concentrations (Skogerboe et al., 1977b).

4.3 ANALYSIS

The choice of analytical method depends on the nature of the data required, the type of sample being analyzed, the skill of the analyst, and the equipment available. For general determination of elemental lead, atomic absorption spectroscopy is widely used and recommended [C.F.R. (1982) 40:§50]. Optical emission spectrometry (Scott et al., 1976b) and X-ray fluorescence (Stevens et al., 1978) are rapid and inexpensive methods for multi-elemental analyses. X-ray fluorescence can measure lead concentrations reliably to 1 ng/m^3 using samples collected with commercial dichotomous samplers. Other analytical methods have specific advantages appropriate for special studies. Only those analytical techniques receiving widespread current use in lead analysis are described below. More complete reviews are available in the literature (American Public Health Association, 1971; Lovering, 1976; Skogerboe et al., 1977b; National Academy of Sciences, 1980).

With respect to measuring lead without sampling or laboratory contamination, several investigators have shown that the magnitude of the problem is quite large (Patterson and Settle, 1976; Patterson et al., 1976; Pierce et al., 1976; Patterson, 1983; Skogerboe, 1982). It appears that the problem may be caused by failure to control the blank or by failure to standardize instrument operation (Patterson, 1983; Skogerboe, 1982). The laboratory atmosphere, collecting containers, and the labware used may be primary contributors to the lead blank problem (Murphy, 1976; Patterson, 1983; Skogerboe, 1982). Failure to recognize these and other sources such as reagents and hand contact is very likely to result in the generation of artificially high analytical results. Samples with less than 100 µg Pb should be analyzed in a clean laboratory especially designed for the elimination of lead contamination. Moody (1982) has described the construction and application of such a laboratory at the National Bureau of Standards.

For many analytical techniques, a preconcentration step is recommended. Leyden and Wegschneider (1981) have described several procedures and the associated problems with controlling the analytical blank. There are two steps to preconcentration. The first is the removal of organic matter by dry ashing or wet digestion. The second is the separation of lead from interfering metallic elements by coprecipitation, co-crystallization, solvent extraction of chelate, electro-deposition or passing through a chelating ion exchange resin column. New separation techniques are continuously being evaluated, many of which have application to specific analytical problems. Torsi and Palmisano (1984) have described electrochemical deposition directly on a glassy carbon crucible during atomic absorption spectrometry. Yang and Yeh (1982) have described a polyacrylamide-hydrous-zirconia (PHZ) composite ion exchanger suitable for high phosphate solutions. Corsini et al. (1982) evaluated a macroreticular acrylic ester resin capable of removing free and inorganically bound metal ions directly from aqueous solution without prior chelation.

Occasionally, it is advantageous to automate the sample preparation and preconcentration process. Tyson (1985) has reviewed the use of flow injection analysis techniques specific for atomic absorption spectrometry. Another promising technique involves a flow-injection system in conjunction with an ion-exchange column and flame atomic absorption (Fang et al., 1984a; 1984b). For aqueous samples, preconcentration factors of 50 to 100-fold were achieved while maintaining a sample frequency of 60 samples per hour.

The application of these and other new techniques can be expected to shed further light on the chemistry and biological availability of lead in natural systems.

4.3.1 Atomic Absorption Spectroscopy (AAS)

Atomic absorption spectroscopy (AAS) is a widely accepted method for the measurement of lead in environmental sampling (Skogerboe et al., 1977b). A variety of lead studies using AAS have been reported (Kometani et al., 1972; Zoller et al., 1974; Huntzicker et al., 1975; Scott et al., 1976b; Lester et al., 1977; Hirao et al., 1979; Compton and Thomas, 1980; Bertenshaw and Gelsthorpe, 1981).

The lead atoms in the sample must be vaporized either in a precisely controlled flame or in a furnace. Furnace systems in AAS offer high sensitivity as well as the ability to analyze small samples (Lester et al., 1977; Rouseff and Ting, 1980; Stein et al., 1980; Bertenshaw and Gelsthorpe, 1981). These enhanced capabilities are offset in part by greater difficulty in analytical calibration and by loss of analytical precision.

Pachuta and Love (1980) collected particles on cellulose acetate filters. Disks (0.5 cm^2) were punched from these filters and analyzed by insertion of the nichrome cups containing the disks into a flame. Another application involves the use of graphite cups as particle filters with the subsequent analysis of the cups directly in the furnace system (Seeley and Skogerboe, 1974; Torsi et al., 1981). These two procedures offer the ability to determine particulate lead directly with minimal sample handling.

In an analysis using AAS and hi-vol samplers, atmospheric concentrations of lead were found to be 0.076 ng/m^3 at the South Pole (Maenhaut et al., 1979). Lead analyses of 995 particulate samples from the NASN were accomplished by AAS with an indicated precision of 11 percent (Scott et al., 1976a; see also Section 7.2.1.1). More specialized AAS methods have been described for the determination of tetraalkyl lead compounds in water and fish tissue (Chau et al., 1979) and in air (Birnie and Noden, 1980; Rohbock et al., 1980).

Atomic absorption requires as much care as other techniques to obtain highly precise data. Background absorption, chemical interference, background light loss, and other factors can cause errors. A major problem with AAS is that untrained operators use it in many laboratories without adequate quality control.

Techniques for AAS are still evolving. An alternative to the graphite furnace, evaluated by Jin and Taga (1982), uses a heated quartz tube through which the metal ion in gaseous hydride form flows continuously. Sensitivities were 1 - 3 ng/g for lead. The technique is similar to the hydride generators used for mercury, arsenic, and selenium. Other nonflame atomization systems, electrodeless discharge lamps, and other equipment refinements and technique developments have been reported (Horlick, 1982). A promising technique for the analysis of samples with high salt content has been developed by Olsen et al. (1983) using flow injection analysis. In an automated system, these authors reported a detection limit of 10 ng/g in seawater while analyzing 30 to 60 samples per hour. This sensitivity is not as low as AAS with a graphite furnace, so the technique would not improve the analysis of air samples without further refinement.

4.3.2 Emission Spectroscopy

Optical emission spectroscopy is based on the measurement of the light emitted by elements when they are excited in an appropriate energy medium. The technique has been used to determine the lead content of soils, rocks, and minerals at the 5 - 10 µg/g level with a relative standard deviation of 5 - 10 percent (Jolly, 1963); this method has also been applied to the analysis of a large number of air samples (Scott et al., 1976b; Sugimae and Skogerboe, 1978). The primary advantage of this method is that it allows simultaneous measurement of a large number of elements in a small sample (Ward and Fishman, 1976).

In a study of environmental contamination by automotive lead, sampling times were shortened by using a sampling technique in which lead-free porous graphite was used both as the filter medium and as the electrode in the spectrometer (Copeland et al., 1973; Seeley and Skogerboe, 1974). Lead concentrations of 1 - 10 µg/m^3 were detected after a half-hour flow at 800 to 1200 ml/min through the filter.

Scott et al. (1976a) analyzed composited particulate samples obtained with hi-vol samplers for 24 elements, including lead, using a direct reading emission spectrometer. Over 1000 samples collected by the NASN in 1970 were analyzed. Careful consideration of accuracy and precision led to the conclusion that optical emission spectroscopy is a rapid and practical technique for particle analysis.

More recent activities have focused attention on the inductively coupled plasma (ICP) system as a valuable means of excitation and analysis (Garbarino and Taylor, 1979; Winge et al., 1977). The ICP system offers a higher degree of sensitivity with less analytical interference than is typical of many of the other emission spectroscopic systems. Optical emission methods are inefficient when used for analysis of a single element, since the equipment is expensive and a high level of operator training is required. This problem is largely offset when analysis for several elements is required, as is often the case for atmospheric aerosols. However, the ICP procedure does not provide the sensitivity required for determining the levels of lead in foods (Jones and Boyer, 1978; Jones et al., 1982).

4.3.3 X-Ray Fluorescence (XRF)

X-ray emissions that characterize the elemental content of a sample also occur when atoms are irradiated at sufficient energy to excite an inner-shell electron (Hammerle and Pierson, 1975; Jaklevic et al., 1973; Skogerboe et al., 1977b; Stevens et al., 1978). This fluorescence allows simultaneous identification of a range of elements including lead.

X-ray fluorescence may require a high-energy irradiation source. But with the X-ray tubes coupled with fluorescers (Jaklevic et al., 1973; Dzubay and Stevens, 1975; Paciga and Jervis, 1976) very little energy is transmitted to the sample; thus sample degradation is kept to a minimum (Shaw et al., 1980). Electron beams (McKinley et al., 1966) and radioactive isotope sources (Kneip and Laurer 1972) have been used extensively as energy sources for XRF analysis (Birks et al., 1971; Birks, 1972). To reduce background interference, secondary fluorescers have been employed (Birks et al., 1971; Dzubay and Stevens, 1975). The fluorescent X-ray emission from the sample may be analyzed with a crystal monochromator and detected with scintillation or proportional counters, or with low-temperature semiconductor detectors that discriminate the energy of the fluorescence. The latter technique requires a very low level of excitation (Dzubay and Stevens, 1975; Toussaint and Boniforti, 1979).

X-ray emission induced by charged-particle excitation (proton-induced X-ray emission or PIXE) offers an attractive alternative to the more common techniques (Barfoot et al., 1979; Hardy et al., 1976; Johansson et al., 1970). The potential of heavy-particle bombardment for excitation was demonstrated by Johansson et al. (1970), who reported an interference-free signal in the picogram (10^{-12} g) range. The excellent capability of accelerator beams for X-ray emission analysis is partially due to the relatively low background radiation associated with the excitation. The high particle fluxes obtainable from accelerators also contribute to the sensitivity of the PIXE method. Literature reviews (Folkmann et al., 1974; Gilfrich et al., 1973; Herman et al., 1973; Walter et al., 1974) on approaches to X-ray elemental analysis agree that protons of a few MeV energy provide a preferred combination for high sensitivity analysis under conditions less subject to matrix interference effects. As a result of this premise, a system designed for routine analysis has been described (Johansson et al., 1975) and papers involving the use of PIXE for aerosol analysis have appeared (Hardy et al., 1976; Johansson et al., 1975). The use of radionuclides to excite X-ray fluorescence and to determine lead in airborne particles has also been described (Havránek and Bumbálová, 1981; Havránek et al., 1980).

X-radiation is the basis of the electron microprobe method of analysis. When an intense electron beam is incident on a sample, it produces several forms of radiation, including X-rays, whose wavelengths depend on the elements present in the material and whose intensities depend on the relative quantities of these elements. An electron beam that gives a spot size as small as 0.2 μm is possible. The microprobe is often incorporated in a scanning electron microscope that allows precise location of the beam and comparison of the sample morphology with its elemental composition. Under ideal conditions, the analysis is quantitative, with an accuracy of a few percent. The mass of the analyzed element may range from 10^{-14} to 10^{-16} g (McKinley et al., 1966).

Electron microprobe analysis is not a widely applicable monitoring method. It requires expensive equipment, complex sample preparation procedures, and a highly trained operator. The method is unique, however, in providing compositional information on individual lead particles, thus permitting the study of dynamic chemical changes and perhaps allowing improved source identification.

Advantages of X-ray fluorescence methods include the ability to detect a variety of elements, the ability to analyze with little or no sample preparation, low detection limits (2 ng Pb/m^3) and the availability of automated analytical equipment. Disadvantages are that the X-ray analysis requires liquid nitrogen (e.g., for energy-dispersive models) and highly trained analysts. The detection limit for lead is approximately 9 ng/cm^2 of filter area (Jaklevic and Walter, 1977), which is well below the quantity obtained in normal sampling periods with the dichotomous sampler (Dzubay and Stevens, 1975).

4.3.4 Isotope Dilution Mass Spectrometry (IDMS)

Isotope dilution mass spectrometry (IDMS) is an absolute measurement technique. It serves as the standard to which other analytical techniques are compared. No other techniques serve more reliably as a comparative reference. Its use for analyses at subnanogram concentrations of lead and in a variety of sample types has been reported (Chow et al., 1969, 1974; Facchetti and Geiss, 1982; Hirao and Patterson, 1974; Murozumi et al., 1969; Patterson et al., 1976; Rabinowitz et al., 1973).

The isotopic composition of lead peculiar to various ore bodies and crustal sources may also be used as a means of tracing the origin of anthropogenic lead. Other examples of IDMS application are found in several reports cited above, and in Rabinowitz and Wetherill (1972), Stacey and Kramers (1975), and Machlan et al. (1976).

4.3.5 Colorimetric Analysis

Colorimetric or spectrophotometric analysis for lead using dithizone (diphenylthiocarbazone) as the reagent has been used for many years (Jolly, 1963; Williams, 1984; Sandell, 1944). It was the primary method recommended by a National Academy of Sciences (1972) report on lead, and the basis for the tentative method of testing for lead in the atmosphere by the American Society for Testing and Materials (1975b). Prior to the development of the IDMS method, colorimetric analysis served as the reference by which other methods were tested.

The procedures for the colorimetric analysis require a skilled analyst. The ASTM conducted a collaborative test of the method (Foster et al., 1975) and concluded that the procedure gave satisfactory precision in the determination of particulate lead in the atmosphere. In addition, the required apparatus is simple and relatively inexpensive, the absorption is linearly related to the lead concentration, large samples can be used, the method is easily sensitive to a few micrograms of lead, and interferences can be removed (Skogerboe et al., 1977b). Realization of these advantages depends on meticulous attention to the procedures and reagents.

4.3.6 Electrochemical Methods: Anodic Stripping Voltammetry (ASV), Differential Pulse Polarography (DPP)

Analytical methods based on electrochemical phenomena are found in a variety of forms (Sawyer and Roberts, 1974; Willard et al., 1974). They are characterized by a high degree of sensitivity, selectivity, and accuracy derived from the relationship between current, charge, potential, and time for electrolytic reactions in solutions. The electrochemistry of lead is based primarily on Pb(II), which behaves reversibly in ionic solutions having a reduction potential near -0.4 volt versus the standard calomel electrode (Skogerboe et al., 1977b). Two electrochemical methods generally offer sufficient analytical sensitivity for most lead measurement problems. Differential pulse polarography (DPP) relies on the measurement of the faradaic current for lead as the voltage is scanned while compensating for the nonfaradaic (background) current produced (McDonnell, 1981). Anodic stripping voltammetry (ASV) is a two step process in which the lead is preconcentrated onto a mercury electrode by an extended but selected period of reduction. After the reduction step, the potential is scanned either linearly or by differential pulse to oxidize the lead and allow measurement of the oxidation (stripping) current. The preconcentration step allows development of enhanced analytical signals; when used in combination with the differential pulse method, lead concentrations at the subnanogram level can be measured (Florence, 1980).

The ASV method has been widely applied to the analysis of atmospheric lead (Harrison et al., 1971; Khandekar et al., 1981; MacLeod and Lee, 1973). Landy (1980) has shown the applicability to the determination of Cd, Cu, Pb, and Zn in Antarctic snow, while others have analyzed rain water (Nguyen et al., 1979; Nürnberg, 1984a; 1984b) and snow samples (Nguyen et al., 1979). Green et al. (1981) have used the method to determine Cd, Cu, and Pb in sea water. The ASV determination of Cd, Cu, Pb, and Zn in foods has been described (Jones et al., 1977; Capar et al., 1982; Mannino, 1982, 1983; Satzger et al., 1982), and the general accuracy of the method summarized by Holak (1980). An ASV method for lead and cadmium in foods has been collaboratively studied and has been adopted as an official method by the Association of Official Analytical Chemists (Capar et al., 1982; Williams, 1984). Current practice with commercially available equipment allows lead analysis at subnanogram concentrations with precision at the 5 to 10 percent level on a routine basis (Skogerboe et al., 1977b). New developments center around the use of microcomputers in controlling the stripping voltage (Kryger, 1981) and conformational modifications of the electrode (Brihaye and Duyckaerts, 1982, 1983). Wang et al. (1983) applied flow-injection techniques to anodic stripping voltammetry to achieve a rate of ten samples per hour.

4.3.7 Methods for Compound Analysis

The majority of analytical methods are restricted to measurement of total lead and cannot directly identify the various compounds of lead. The electron microprobe and other X-ray fluorescence methods provide approximate data on compounds on the basis of the ratios of elements present (Ter Haar and Bayard, 1971). Gas chromatography (GC) using the electron capture detector has been demonstrated to be useful for organolead compounds (Shapiro and Frey, 1968). The use of atomic absorption as the GC detector for organolead compounds has been described by DeJonghe et al. (1981) and Hewitt and Harrison (1985), while a plasma emission detector has been used by Estes et al. (1981). In addition, Messman and Rains (1981) have used liquid chromatography with an atomic absorption detector to measure organolead compounds. Mass spectrometry may also be used with GC (Mykytiuk et al., 1980).

Powder X-ray diffraction techniques have been applied to the identification of lead compounds in soils by Olson and Skogerboe (1975) and by Linton et al. (1980). X-ray diffraction techniques were used (Harrison and Perry, 1977; Foster and Lott, 1980; Jaklevic et al., 1981) to identify lead compounds collected on air filters.

4.4 CONCLUSIONS

To monitor lead particles in air, collection with the hi-vol and dichotomous samplers and analysis by atomic absorption spectrometry and X-ray fluorescence methods have emerged as the most widely used methods. Sampling with the hi-vol has inherent biases in sampling large particles and does not provide for fractionation of the particles according to size, nor does it allow determination of the gaseous (organic) concentrations. Sampling with a dichotomous sampler provides size information but does not permit measurement of gaseous lead. The size distribution of lead aerosol particles is important in considering inhalable particulate matter. X-ray fluorescence and optical emission spectroscopy are applicable to multi-element analysis. Other analytical techniques find application for specific purposes.

There is no routine monitoring program in the United States for ambient concentrations of gaseous lead. Such measurements would require the addition of a chemical scrubber to the particulate sampling device, a procedure that is used only under special circumstances. Discussion of the concentrations of gaseous lead are found in Section 6.3.2.

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5. SOURCES AND EMISSIONS

5.1 HISTORICAL PERSPECTIVE

The history of global lead emissions has been assembled from chronological records of deposition in polar snow strata, marine and freshwater sediments, coral skeleton bands, and the annual rings of trees. These records are important for two reasons. They aid in establishing natural background levels of lead in air, soils, plants, animals, and humans. They also place current trends in atmospheric lead concentrations in the perspective of historical changes. Most chronological records document the sudden increase in atmospheric lead at the time of the industrial revolution, and a later burst starting in the 1920's when lead-alkyls were first added to gasoline.

Tree ring analyses are not likely to show the detailed year-by-year chronological record of atmospheric lead increases. In situations where ring-porous trees (species that retain the nutrient solution only in the most recent annual rings) grow in heavily polluted areas where soil lead has increased 100-fold, significant increases in the lead content of tree rings over the last several decades have been documented. Rolfe (1974) found 4-fold increases in both rural and urban tree rings using pooled samples from the period of 1910-20 compared to samples from the period from 1963-73. Symeonides (1979) found a 2-fold increase during a comparable interval at a high lead site but no increase at a low lead site. Baes and Ragsdale (1981) found significant post-1930 increases in oak (Quercus) and hickory (Carya) with high lead exposure, but only in hickory with low lead exposure. Dodge and Gilbert (1984) reported a chronological increase in lead deposited in the annual bands of coral skeletons near St. Croix, U.S. Virgin Islands. The 2-fold increase from 1950 to 1980 in the coral at the relatively unpolluted site appeared to reflect regional or global deposition.

Pond sediment analyses (Shirahata et al., 1980) have shown a 20-fold increase in lead deposition during the last 150 years in the western United States (Figure 5-1), documenting not only the increasing use of lead since the beginning of the industrial revolution in that region, but also the relative fraction of natural vs. anthropogenic lead inputs. Other studies have shown a similar magnitude of increasing deposition in freshwater sediments (Christensen and Chien, 1981; Galloway and Likens, 1979; Edgington and Robbins, 1976; Dominik et al., 1984; Wong et al., 1984), and marine sediments (Ng and Patterson, 1982). The pond and marine sediments of Shirahata et al. (1980) and Ng and Patterson (1982) also document the shift in isotopic composition caused by the recent opening of the New Lead Belt in Missouri (see Section 5.3.3.2) where the ore body has an isotopic composition substantially different from other ore bodies of the world.

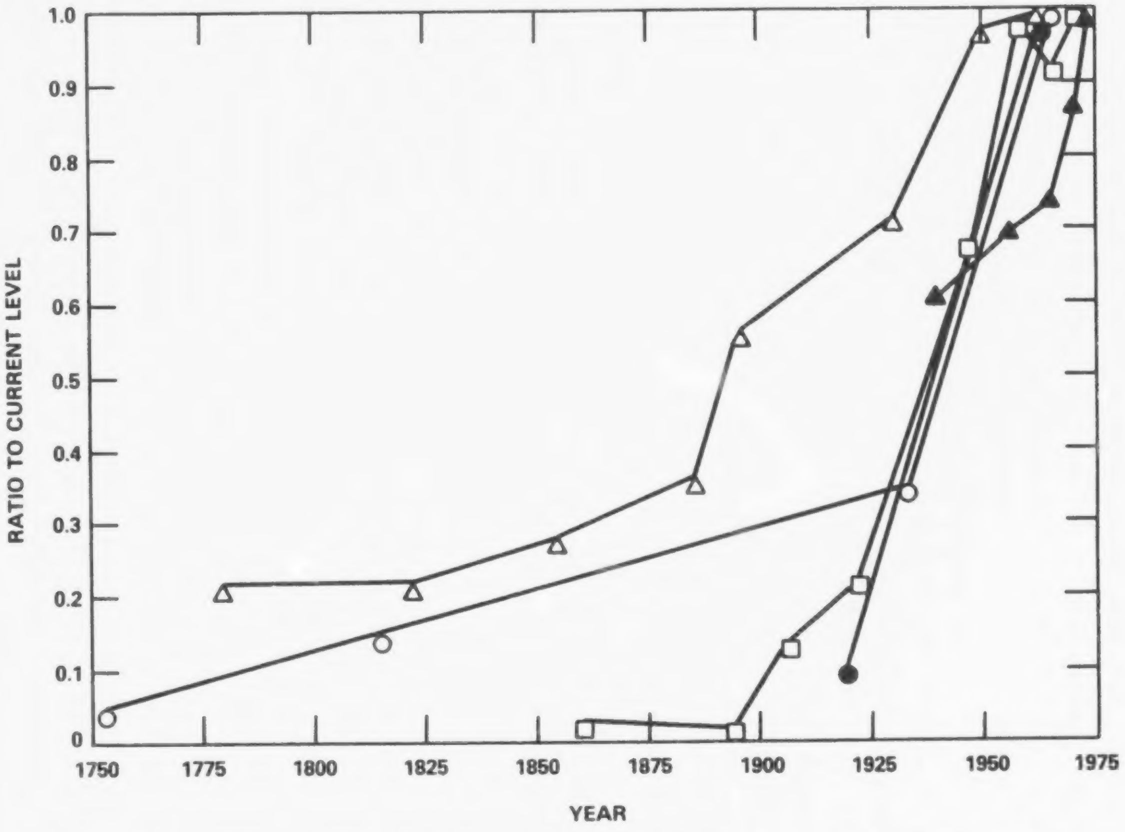

Figure 5-1. Chronological record of the relative increase of lead in snow strata, pond and lake sediments, marine sediments, and tree rings. The data are expressed as a ratio of the latest year of the record and should not be interpreted to extend back in time to natural or uncontaminated levels of lead concentration.

Source: Adapted from Murozumi et al. (1969) (○), Shirahata et al. (1980) (□), Edgington and Robbins (1976) (△), Ng and Patterson (1982) (▲), and Rolfe (1974) (●).

Perhaps the best and certainly the most controversial chronological record is that of the polar ice strata of Murozumi et al. (1969), which extends nearly three thousand years back in time (Figure 5-1). In a comprehensive review of chronological studies of global pollution in polar snow and ice, Wolff and Peel (1985) concluded that, although a few samples in the Greenland study of Murozumi et al. (1969) may have been contaminated, the results are valid and have been confirmed by later studies (Ng and Patterson, 1981). Intermediate studies that reported much higher concentrations were probably erroneous.

In Antarctica, lead concentrations in snow and ice are about one tenth of the values from polar regions in the northern hemisphere. This phenomenon has been attributed to the restricted interchange in the atmospheric circulation patterns between the northern and southern hemispheres, and to the fact that 90 percent of the global industrial activity occurs in the northern hemisphere (Wolff and Peel, 1985). Recent studies by Wolff and Peel (1985) confirmed the values of 5 pg Pb/g snow found by Boutron and Patterson (1983), repudiating many previous studies that reported higher values.

It is likely that prehistoric concentrations of lead in snow and ice of Greenland and Antarctica were a maximum of 1.4 and 1.2 pg/g (Ng and Patterson, 1983; Boutron and Patterson, 1983), while present concentrations are 200 pg/p in Greenland (Murozumi et al., 1969) and 5-6 pg/g in Antarctica (Boutron and Patterson, 1983). Data for Antarctica agree with atmospheric measurements of Maenhaut et al. (1979), who found air concentrations of 0.000076 $\mu g/m^3$ suggested by Patterson (1980) and Servant (1982) as the natural lead concentration in the atmosphere.

In summary, it is likely that atmospheric lead emissions have increased 2000-fold since the pre-Roman era, that even at this early time the atmosphere may have been contaminated by a factor of three over natural levels (Murozumi et al. 1969), and that global atmospheric concentrations have increased dramatically since the 1920's.

The history of global emissions may also be determined from total production of lead, if the amounts of lead released to the atmosphere during the smelting process, released during industrial consumption, and emitted from non-lead sources are known. The historical picture of lead production has been pieced together from many sources by Settle and Patterson (1980) (Figure 5-2). They used records of accumulated silver stocks to estimate the lead production needed to support coin production. Until the industrial revolution, lead production was determined largely by the ability or desire to mine lead for its silver content. Since that time, lead has been used as an industrial product in its own right, and efforts to improve smelter efficiency, including control of stack emissions and fugitive dusts, have made lead production more economical. This improved efficiency is not reflected in the chronological record because of atmospheric emissions of lead from many other anthropogenic sources, especially gasoline combustion (see Section 5.3.3). From this knowledge of the chronological

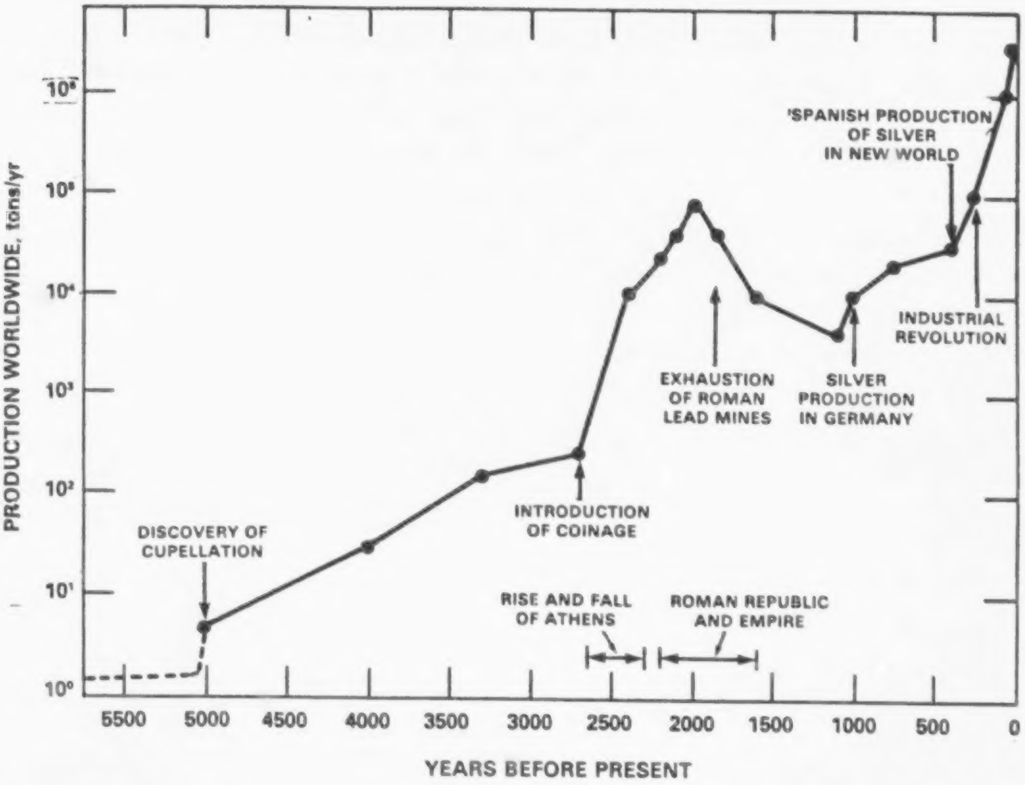

Figure 5-2. The global lead production has changed historically in response to major economic and political events. Increases in lead production (note log scale) correspond approximately to historical increases in lead emissions shown in Figure 5-1.

Source: Adapted from Settle and Patterson (1980).

record, it is possible to sort out contemporary anthropogenic emissions from natural sources of atmospheric lead.

5.2 NATURAL SOURCES

Lead enters the biosphere from lead-bearing minerals in the lithosphere through both natural and man-made processes. Measurements of soil materials taken at 20-cm depths in the continental United States (Lovering, 1976; Shacklette et al. 1971) show a median lead concentration of 15 - 16 µg Pb/g soil. Ninety-five percent of these measurements show 30 µg/g of lead or less, with a maximum sample concentration of 700 µg/g.

In natural processes, lead is first incorporated in soil in the active root zone, from which it may be absorbed by plants, leached into surface waters, or eroded into windborne dusts (National Academy of Sciences, 1980; Chamberlain, 1970; Patterson, 1965; Chow and Patterson, 1962).

Natural emissions of lead from volcanoes have been estimated by Nriagu (1979) to be 6400 metric tons (t)/year based on enrichment over crustal abundance. That is, 10×10^9 kg/year of volcanic dust are produced, with an average lead concentration of 640 μg/g, or 40 times the crustal abundance of 16 μg/g. The enrichment factor is based on Lepel et al. (1978), who measured lead in the plume of the Augustine volcano in Alaska. Settle and Patterson (1980) have calculated emissions of only 1 t/year, based on a measured Pb/S ratio of 2×10^{-7} (Buat-Menard and Arnold, 1978), and estimated sulfur emissions of 6×10^6 t/year. The estimate of Settle and Patterson (1980) is more direct, and perhaps more reliable, because it depends on estimates of sulfur emissions rather than total volcanic dust.

Calculations of natural contributions using geochemical information indicate that natural sources contribute a relatively small amount of lead to the atmosphere. For example, if the typical 25 - 40 μg/m^3 of rural airborne particulate matter consisted solely of wind-entrained soils containing 15 μg/g (and rarely more than 30 μg of lead/g), as cited above, then the natural contribution to airborne lead would range from 0.0004 to 0.0012 μg/m^3. It has been estimated from geochemical evidence that the natural particulate lead level is less than 0.0005 μg/m^3 (National Academy of Sciences, 1980; United Kingdom Department of the Environment, 1974). In fact, levels as low as 0.000076 μg/m^3 have been measured at the South Pole in Anarctica (Maenhaut et al., 1979). In contrast, lead concentrations in urban suspended particulate matter may be as high as 6 μg/m^3 (Akland, 1976; U.S. Environmental Protection Agency, 1979, 1978). Most of this urban particulate lead stems from manmade sources.

5.3 MANMADE SOURCES

5.3.1 Production

Lead occupies an important position in the U.S. economy, ranking fifth among all metals in tonnage used. Approximately 85 percent of the primary lead produced in this country is from native mines; it is often associated with minor amounts of zinc, cadmium, copper, bismuth, gold, silver, and other minerals (U.S. Bureau of Mines, 1975). Missouri lead ore deposits account for approximately 80 to 90 percent of the domestic production. Approximately 40 to 50 percent of annual lead production is recovered and eventually recycled.

.2 Utilization

The 1971-1982 uses of lead are listed by major product category in Table 5-1 (U.S. Bureau of Mines, 1972-1984). Total utilization averaged approximately 1.29 x 10^6 t/yr over the 12-year period, with storage batteries and gasoline additives accounting for ~70 percent of total use. The gasoline antiknock compounds listed in Table 5-1 include additives for both domestic and import markets. The additive fraction of total lead utilization has decreased from greater than 18 percent in 1971-1973 to less than 9.5 percent in 1981. Certain products, especially batteries, cables, plumbing, weights, and ballast, contain lead that is economically recoverable as secondary lead. This reserve of lead in use is estimated at 3.8 million metric tons. Of the one million metric tons of lead used in commercial products annually, 0.5 to 0.8 million tons are recovered. Lead used in pigments, gasoline additives, ammunition, foil, solder, and steel products is widely dispersed and therefore is largely unrecoverable.

5.3.3 Emissions

Lead or its compounds may enter the environment at any point during mining, smelting, processing, use, recycling, or disposal. Estimates of the dispersal of lead emissions into the environment by principal sources indicate that the atmosphere is the major initial recipient. Estimated lead emissions to the atmosphere are shown in Table 5-2. Mobile and stationary sources of lead emissions, although found throughout the nation, tend to be concentrated in areas of high population density, with the exception of smelters. Figure 5-3 shows the approximate locations of major lead mines, primary and secondary smelters and refineries, and alkyl lead plants (International Lead Zinc Research Organization, 1982).

5.3.3.1 Mobile Sources. The majority of lead compounds found in the atmosphere result from leaded gasoline combustion. Several reports indicate that transportation sources, which include light-duty, heavy-duty, and off-highway vehicles, contribute over 80 percent of the total atmospheric lead (Nationwide [lead] emissions report, 1980, 1979; U.S. Environmental Protection Agency, 1977). Other mobile sources, including aviation use of leaded gasoline and diesel and jet fuel combustion, contribute insignificant lead emissions to the atmosphere. The detailed emissions inventory in Table 5-2 shows that 89 percent of the lead emissions in the United States are from gasoline combustion. Cass and McRae (1983) assembled emissions inventory data on the Los Angeles Basin and determined that 83 percent of the fine particle emissions originated from highway vehicles. Lead is added to gasoline as an antiknock additive to enhance engine performance in the form of two tetralkyl lead compounds, tetraethyl and tetramethyl lead (see Section 3.4). Lead is emitted from vehicles primarily in the form of inorganic particles, although a very small fraction (<10 percent) of lead emissions are released as volatile organic compounds, i.e., lead alkyls (see Section 6.3.2)

TABLE 5-1. U.S. UTILIZATION OF LEAD BY PRODUCT CATEGORY (1971-1980)
(metric tons/yr)

Product category	1971	1972	1973	1974	1975	1976	1977	1978	1979	1980	1981	1982	1983	1984
Storage batteries	616,581	661,740	697,888	772,656	634,368	746,085	858,099	879,274	814,332	645,357	770,152	704,323	806,999	865, 547
Gasoline antiknock additives[a]	239,666	252,545	248.890	227,847	189,369	217,508	211,296	178,473	186,945	127,903	111,367	119,234	89,118	78,933
Pigments and ceramics	73,701	80,917	98,651	105,405	71,718	95,792	90,704	91,642	90,790	78,430	80,165	60,866	68,694	76,808
Ammunition	79,423	76,822	73,091	78,991	68,098	66,659	62,043	55,776	53,236	48,662	49,514	44,237	43,697	47,828
Solder	63,502	64,659	65,095	60,116	52,011	57,448	58,320	68,390	54,278	41,366	29,705	28,500	28,490	24,441
Cable coverings	47,998	41,659	39,006	39,387	20,044	14,452	13,705	13,851	16,393	13,408	12,072	15,181	10,505	12,270
Caulking lead	27,204	20,392	18,192	17,903	12,966	11,317	8,725	9,909	8,017	5,684	5,522	4,056	3,572	3,966
Pipe and sheet lead	41,523	37,592	40,529	34,238	35,456	34,680	30,861	23,105	27,618	28,393	28,184	23,838	27,261	28,323
Type metal	18,876	18,089	19,883	18,608	14,703	13,614	11,395	10,795	10,019	8,997	7,838	2,766	2,540	2,162
Brass and bronze	18,180	17,963	20,621	20,172	12,157	14,207	15,148	16,502	18,748	13,981	13,306	11,352	10,980	6,954
Bearing metals	14,771	14,435	14,201	13,250	11,051	11,851	10,873	9,510	9,630	7,808	6,922	6,133	5,844	4,677
Other	56,958	63,124	61,019	62,106	54,524	68,181	64,328	75,517	68,329	50,314	52,354	54,922	50,887	55,124
TOTAL	1,298,383	1,349,846	1,397,876	1,450,679	1,176,465	1,351,794	1,435,497	1,432,744	1,358,335	1,070,303	1,167,101	1,075,408	1,148,487	1,207,033

[a] Includes additives for both domestic and export markets.

Source: U.S. Bureau of Mines (1972-1984).

TABLE 5-2. ESTIMATED ANTHROPOGENIC LEAD EMISSIONS TO THE ATMOSPHERE FOR THE UNITED STATES, 1984

Source Category	Annual (1984) emissions, (t/yr)	Percentage of total U.S. emissions
Gasoline combustion	34,881	89.4%
Waste oil combustion	781	2.0
Solid waste disposal	352	0.9
Coal combustion	265	0.7
Oil combustion	115	0.3
Gray iron production	54	0.1
Iron and steel production	427	1.1
Secondary lead smelting	278	0.7
Primary copper smelting	29	0.1
Ore crushing and grinding	116	0.3
Primary lead smelting	1150	2.8
Zn smelting	116	0.3
Other metallurgical	11	0.1
Lead alkyl manufacture	224	0.6
Lead acid battery manufacture	112	0.3
Portland cement production	70	0.2
Miscellaneous	35	0.1
Total	39,016[a]	100%

[a] Inventory does not include emissions from exhausting workroom air, burning of lead-painted surfaces, welding of lead-painted steel structures, or weathering of painted surfaces.

Source: Updated from Battye (1983).

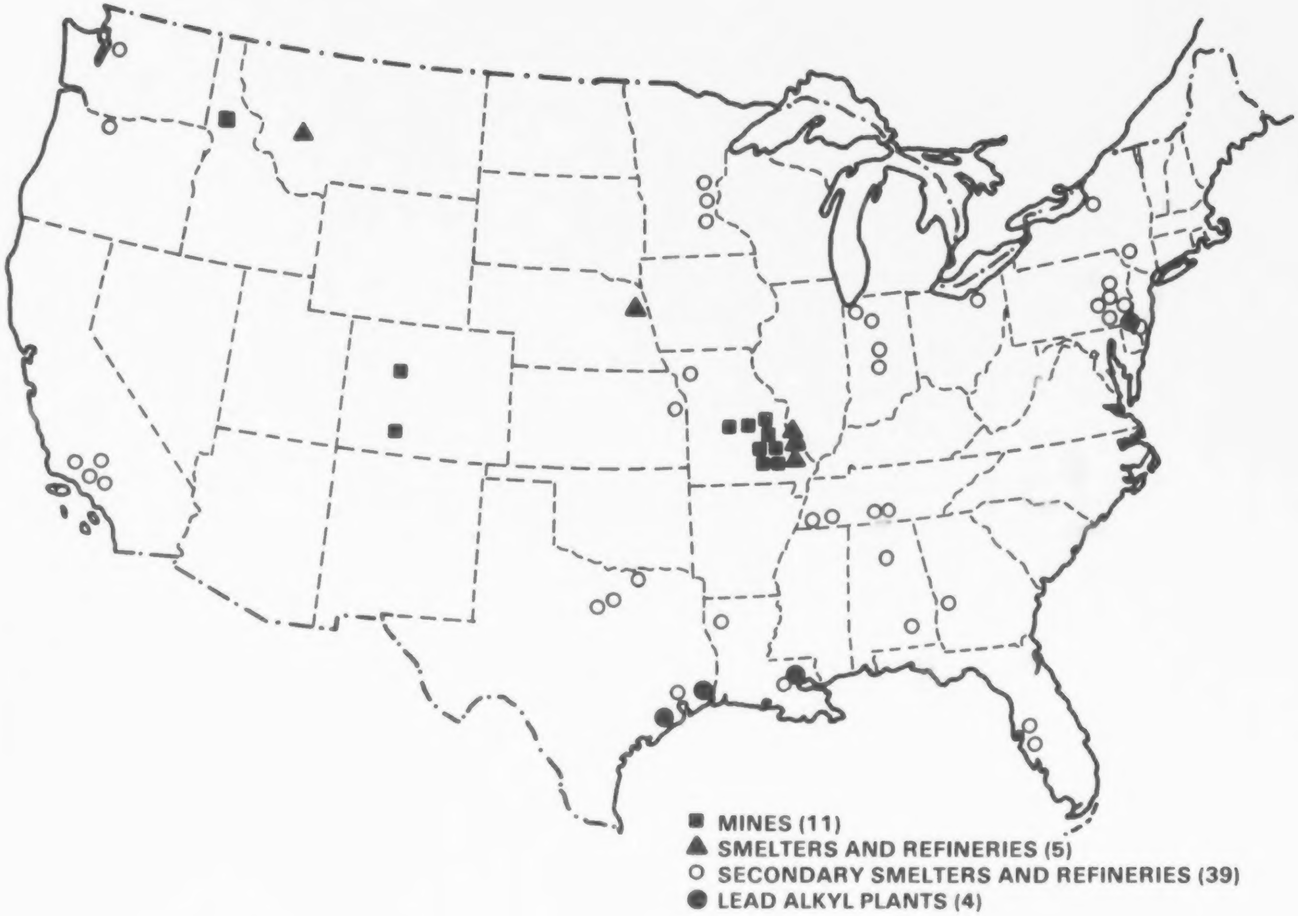
■ MINES (11)
▲ SMELTERS AND REFINERIES (5)
○ SECONDARY SMELTERS AND REFINERIES (39)
● LEAD ALKYL PLANTS (4)

Figure 5-3. Locations of major lead operations in the United States.

Source: International Lead Zinc Research Organization (1985).

Commercial lead antiknock additives of all types contain halogens designated as scavengers that serve to reduce the accumulation of decomposition products of the lead alkyls in certain critical areas of the engine combustion chamber. The most commonly used additive package contains enough ethylene dibromide to tie up all of the lead as $PbBr_2$, and enough ethylene dichloride to tie up 1.5 times the amount of lead as $PbCl_2$.

The factors which affect both the rate of particulate lead emissions and the physicochemical properties of the emissions are: lead content of the fuel, other additives, vehicle fuel economy, the driving speed or conditions, and type of vehicle, as well as design parameters, maintenance, and ages of the engine, exhaust, and emission control systems. The major types of vehicles are light-duty (predominantly cars) and heavy-duty (trucks and buses). The important properties of the particulate emissions include the total amount emitted, the size distribution of the particles, and the chemical composition of these particles as a function of particle size. The most commonly used index of particle size is the mass median aerodynamic diameter (MMAD), which is defined as the point in the size distribution of particles such that half the mass lies on either side of the MMAD value (National Air Pollution Control Administration, 1969). Table 5-3 summarizes a recent study estimating the particulate emission rates and particle composition for light-duty vehicles operated on a leaded fuel of 1.8 g Pb/gallon (Hare and Black, 1981). Table 5-4 estimates particulate emission rates for heavy-duty vehicles (trucks) operated on a leaded fuel of 1.8 g Pb/gallon (Hare and Black, 1981). The lead content of 1.8 g Pb/gallon was chosen to approximate the lead concentration of leaded gasoline during 1979 (Table 5-5). Another recent study utilizing similar composite emission factors provides estimates of motor vehicle lead emissions for large areas (Provenzano, 1978).

The fate of emitted lead particles depends upon their particle size (see Section 6.3.1). Particles initially formed by condensation of lead compounds in the combustion gases are quite small (well under 0.1 μm in diameter, see Section 6.3.1) (Pierson and Brachaczek, 1983). Particles in this size category are subject to growth by coagulation and, when airborne, can remain suspended in the atmosphere for 7 - 30 days and travel thousands of miles from their original source (Chamberlain et al., 1979). Larger particles are formed as the result of agglomeration of smaller condensation particles and have limited atmospheric lifetimes (Harrison and Laxen, 1981). The largest vehicle-emitted particles, which are greater than 100 μm in diameter, may be formed by materials flaking off from the surfaces of the exhaust system. As indicated in Table 5-3, the estimated mass median equivalent diameter of leaded particles from light-duty vehicles is <0.25 μm, suggesting that such particles with relatively long atmospheric lifetimes have the potential for long-distance transport. Similar values for MMAD in automobile exhausts were found in Britain (0.27 μm) (Chamberlain et al. 1979) and

TABLE 5-3. LIGHT-DUTY VEHICULAR PARTICULATE EMISSIONS*

Rate or property	Data by vehicle category Pre-1970	 1970 & later without catalyst
Exhaust particulate emissions, g/mi (g/km)	0.29 (0.47)	0.13 (0.21)
Particle mass median equivalent diameter, μm	<0.25	<0.25
Percent of particulate mass as:		
Lead (Pb)	22 or greater	36 or greater
Bromine (Br)	11 or greater	18 or greater
Chlorine (Cl)	4 or greater	6 or greater
Trace metals	1	1 or greater
Carbon (C), total	33 or greater	33 or less
Sulfate ($SO_4 2-$)	1.3	1.3 or greater
Soluble organics	~30 or less	~10

*Rate estimates are based on 1.8 g Pb/gal (0.42 g/l) fuel.

Source: Hare and Black (1981).

TABLE 5-4. HEAVY-DUTY VEHICULAR PARTICULATE EMISSIONS*
[g/mi (g/km)]

Heavy-duty category	Particulate emissions by model year Pre-1970	 1970 and later
Medium-duty trucks (6,000 to 10,000 lb)†	0.50 (0.80)	0.40 (0.64)
Heavy-duty trucks (over 10,000 lb)†	0.76 (1.2)	0.60 (0.96)

*Rate estimates are based on 1.8 g Pb/gal (0.42 g/l) fuel.
†Gross vehicular weight.

Source: Hare and Black (1981).

TABLE 5-5. RECENT AND PROJECTED CONSUMPTION OF GASOLINE LEAD

Calendar year	Gasoline volume 10^9 gal Total	Gasoline volume 10^9 gal Leaded	Average lead content g/gal Pooled	Average lead content g/gal Leaded	Total lead 10^3t	Air-lead µg/m^3
1975[a]	102.3	92.5	1.62	1.81	167.4	1.23
1976	107.0	87.0	1.60	1.97	171.4	1.22
1977	113.2	79.7	1.49	2.12	168.9	1.20
1978	115.8	75.0	1.32	2.04	153.0	1.13
1979	111.2	68.1	1.16	1.90	129.4	0.74[c]
1980	110.8	57.5	0.71	1.37	78.8	0.66[c]
1981	102.6	51.0	0.59	1.19	60.7	0.51[c]
1982[b]	98.7	52.5	0.61	1.14	59.9	0.53[c]
1983	102.4	47.5	0.51	1.10	52.3	0.40[c]
1984	105.7	43.8	0.44	1.05	46.0	0.36[c]
1985[d]	100.6	32.2	0.26	0.80	25.8	
1986	100.3	28.8	0.03	0.10	2.9	
1987	100.0	25.6	0.03	0.10	2.6	
1988	99.3	22.4	0.02	0.10	2.2	
1989	99.0	19.2	0.02	0.10	1.9	
1990	99.0	16.4	0.02	0.10	1.6	

[a] Data for the years 1975-1981 are taken from U.S. Environmental Protection Agency (1983).

[b] Data for 1982-1984 are taken from U.S. Environmental Protection Agency (1985).

[c] Data from U.S. EPA (1986), discussed in Chapter 7, are the maximum quarterly average lead levels from a composite of 147 sampling sites. Earlier reports for the period 1975-78 were based on a different, although comparable group of sites.

[d] Data for 1985-1990 are estimates taken from F.R. (1985 March 7).

Italy (0.33 µm) (Facchetti and Geiss, 1982). Particles this small deposit by Brownian diffusion and are generally independent of gravitation (see Section 6.4.1.1).

The size distribution of lead particles is essentially bimodal at the time of exhaust (Pierson and Brachaczek, 1976, 1983) and depends on a number of factors, including the particular driving pattern in which the vehicle is used and its past driving history (Ganley and Springer, 1974; Habibi, 1973, 1970; Ter Haar et al., 1972; Hirschler and Gilbert, 1964; Hirschler et al., 1957). As an overall average, it has been estimated that during the lifetime of the vehicle, approximately 35 percent of the lead contained in the gasoline burned by the vehicle is emitted as small particles (<0.25 µm MMAD), and approximately 40 percent is emitted as larger particles (>10 µm MMAD) (Ter Haar et al., 1972). The remainder of the lead consumed in gasoline combustion is deposited in the engine and exhaust system. Engine deposits are, in part, gradually transferred to the lubricating oil and removed from the vehicle when the oil is changed. A flow chart depicting lead-only emissions per gallon of fuel charged into the engine is shown in Figure 5-4. It is estimated that 10 percent of the lead consumed during combustion is released into the environment via disposal of used lubricating oil (Piver, 1977). In addition, some of the lead deposited in the exhaust system gradually flakes off, is emitted in the exhaust as extremely large particles, and rapidly falls into the streets and roads where it is incorporated into the dust and washed into sewers or onto adjacent soil.

Although the majority (>90 percent on a mass basis) of vehicular lead compounds are emitted as inorganic particles (e.g., PbBrCl), some organolead vapors (e.g., lead alkyls) are also emitted. The largest volume of organolead vapors arises from the manufacture, transport, and handling of leaded gasoline. Such vapors are photoreactive and their presence in local atmospheres is transitory; i.e., the estimated atmospheric half-lives of lead alkyls, under typical summertime conditions, are less than half a day (Nielsen, 1984). Organolead vapors are most likely to occur in occupational settings (e.g., gasoline transport and handling operations, gas stations, parking garages) and have been found to contribute less than 10 percent of the total lead present in the atmosphere (Gibson and Farmer, 1981; National Academy of Sciences, 1972).

The use of lead additives in gasoline, which increased in volume for many years, is now decreasing as automobiles designed to use unleaded fuel constitute the major portion of the fleet (Table 5-1). The decline in the use of leaded fuel is the result of two regulations promulgated by the U.S. Environmental Protection Agency (F.R., 1973, December 6). The first required the availability of unleaded fuel for use in automobiles designed to meet federal emission standards with lead-sensitive emission control devices (e.g., catalytic converters); the second required a reduction or phase-down of the lead content in leaded gasoline. The phase-down schedule of lead in gasoline was modified in 1982 (F.R., 1982, October 29), replacing the 0.5 g/gal standard for the average lead content of all gasoline with a standard of 1.10 g Pb/gal for leaded gasoline alone, and again in 1985 (F.R., 1985, March 7), calling for a reduction to 0.5 g Pb/gal leaded gas by July 1985 and 0.1 g Pb/gal leaded gas by January 1986.

The trend in lead content for U.S. gasolines is shown in Figure 5-5 and Table 5-5. Of the total gasoline pool, which includes both leaded and unleaded fuels, the average lead content has decreased 73 percent, from an average of 1.62 g/gal in 1975 to 0.44 g/gal in 1984 (Table 5-5, Figure 5-5). Accompanying the phase-down of lead in leaded fuel has been the

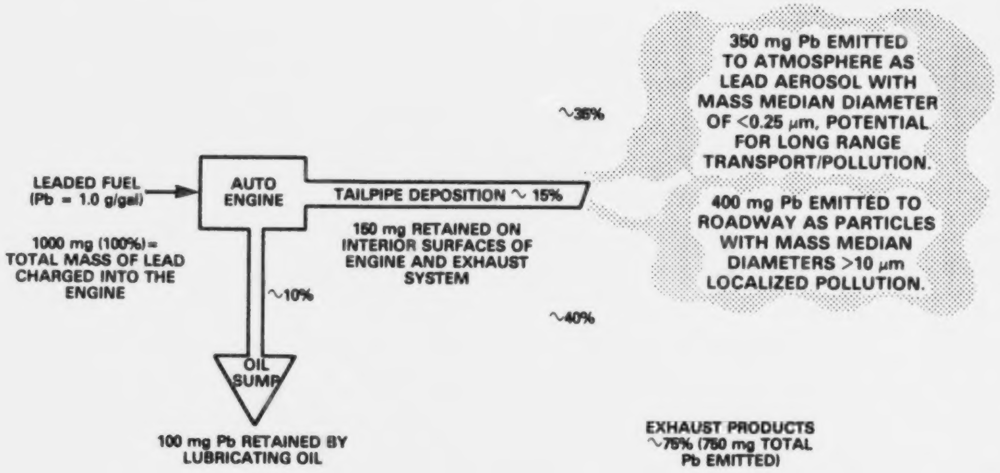
LEADED FUEL (Pb = 1.0 g/gal)
1000 mg (100%) = TOTAL MASS OF LEAD CHARGED INTO THE ENGINE
AUTO ENGINE
TAILPIPE DEPOSITION ~ 15%
150 mg RETAINED ON INTERIOR SURFACES OF ENGINE AND EXHAUST SYSTEM
~10%
OIL SUMP
100 mg Pb RETAINED BY LUBRICATING OIL
~35%
350 mg Pb EMITTED TO ATMOSPHERE AS LEAD AEROSOL WITH MASS MEDIAN DIAMETER OF <0.25 μm, POTENTIAL FOR LONG RANGE TRANSPORT/POLLUTION.
~40%
400 mg Pb EMITTED TO ROADWAY AS PARTICLES WITH MASS MEDIAN DIAMETERS >10 μm LOCALIZED POLLUTION.
EXHAUST PRODUCTS ~75% (750 mg TOTAL Pb EMITTED)

Figure 5-4. Estimated lead-only emissions distribution per gallon of combusted fuel.

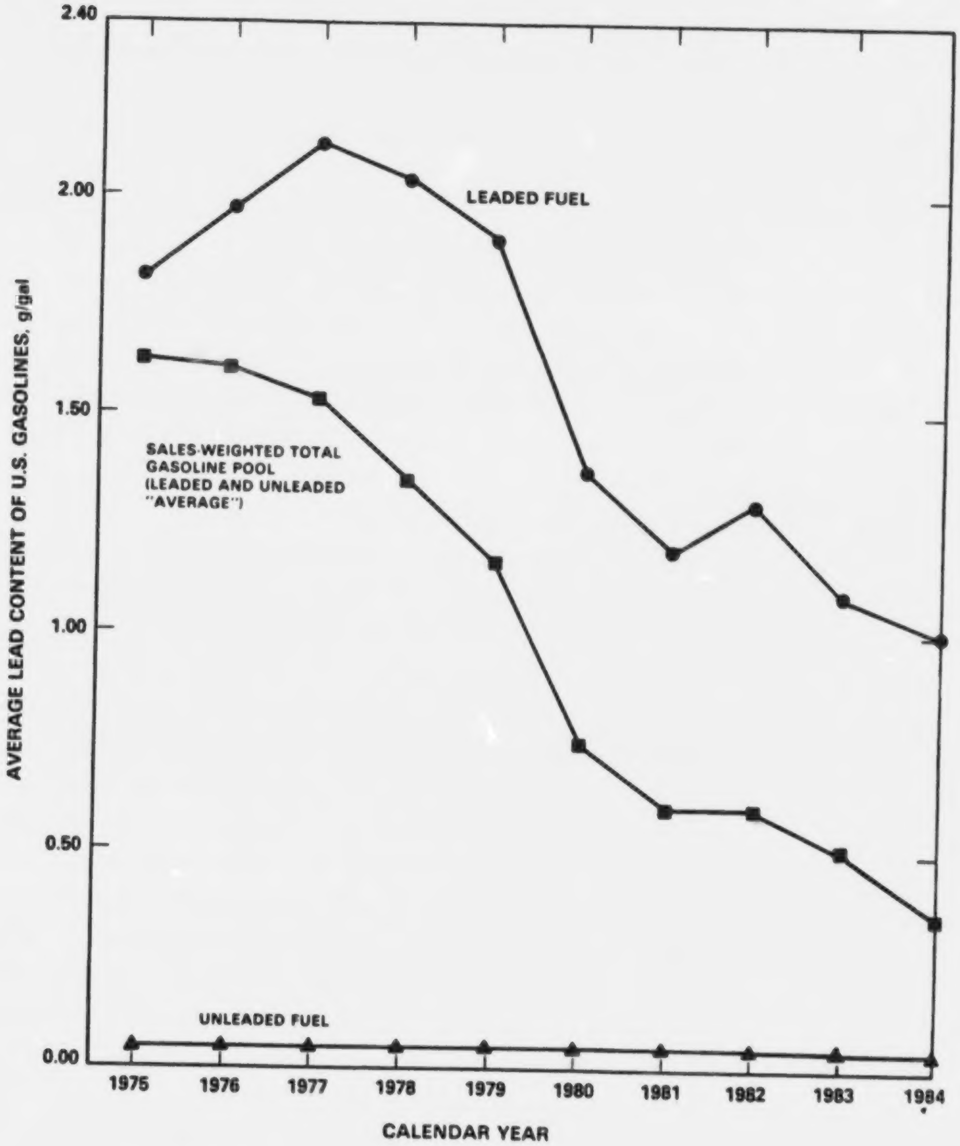

Figure 5-5. Trend in lead content of U.S. gasolines, 1975-1984.

Source: U.S. EPA (1985).

increased consumption of unleaded fuel, from 10 percent of the total gasoline pool in 1975 to 59 percent in 1984 (Table 5-5 and Figure 5-6). Since 1975, when the catalytic converter was introduced by automobile manufacturers for automotive exhaust emissions control, virtually all new passenger cars have been certified on unleaded gasoline (with the exception of a few diesels and a very few leaded-gasoline vehicles).

Data describing the lead consumed in gasoline and average ambient lead levels (composite of maximum quarterly values) vs. calendar year are listed in Table 5-5 and plotted in Figure 5-7. The 1975 through 1979 composite quarterly lead averages are based on 105 lead-monitoring sites, primarily urban. The 1980 through 1984 composite average is based on 147 sites with valid annual data. Between 1975 and 1984, the lead consumed in gasoline decreased 73 percent (from 167,400 to 46,000 metric tons) while the corresponding composite maximum quarterly average of ambient air lead decreased 71 percent (from 1.23 to 0.36 $\mu g/m^3$). This indicates that control of lead in gasoline over the past several years has effected a direct decrease in peak ambient lead concentrations, at least for this group of monitoring sites.

5.3.3.2 Stationary Sources. As shown in Table 5-2 (based on 1984 emission estimates), primary lead smelting, coal combustion, and combustion of waste oil are the principal contributors of lead emissions from stationary sources. Coal-fired electric power stations typically burn 5,000 to 10,000 tons of coal per day. Pulverized coal is mixed with hot air and passed into a burning chamber or boiler, where the mixture is ignited. Some of the unburned residue falls to the bottom of the boiler, where it is removed as 'bottom ash'. The residue that passes through the boiler is called 'fly ash', much of which is removed by electrostatic precipitators and other pollution control devices. In a well-designed system, 99.8 percent of the original inorganic mass of the coal is retained by the system. At 10 g Pb/ton of coal, very little lead would be emitted. However, the remaining 0.2 percent of the coal mass that is emitted from the stack is highly enriched in lead, compared to the original coal. Although data on stack emissions of lead are limited, the concentration of lead in fly ash may provide a reasonable indication of stack lead emissions. Klusek et al. (1983) reported an enrichment of 6.1 between coal and fly ash. On this basis, a typical power plant consuming 10,000 tons of coal per day would emit 1.2 kg Pb/day (10,000 t/day x 0.002 x 6.1 x 10 g Pb/t coal). Turner and Lowry (1983) reported enrichment factors of 17 to 75 for conventional coal-fired power plants in Pennsylvania and New Hampshire.

The manufacture of consumer products such as lead glass, storage batteries, and lead additives for gasoline also contributes significantly to stationary source lead emissions. Since 1970, the quantity of lead emitted from the metallurgical industry has decreased somewhat because of the application of control equipment and the closing of several plants, particularly in the zinc and pyrometallurgical industries.

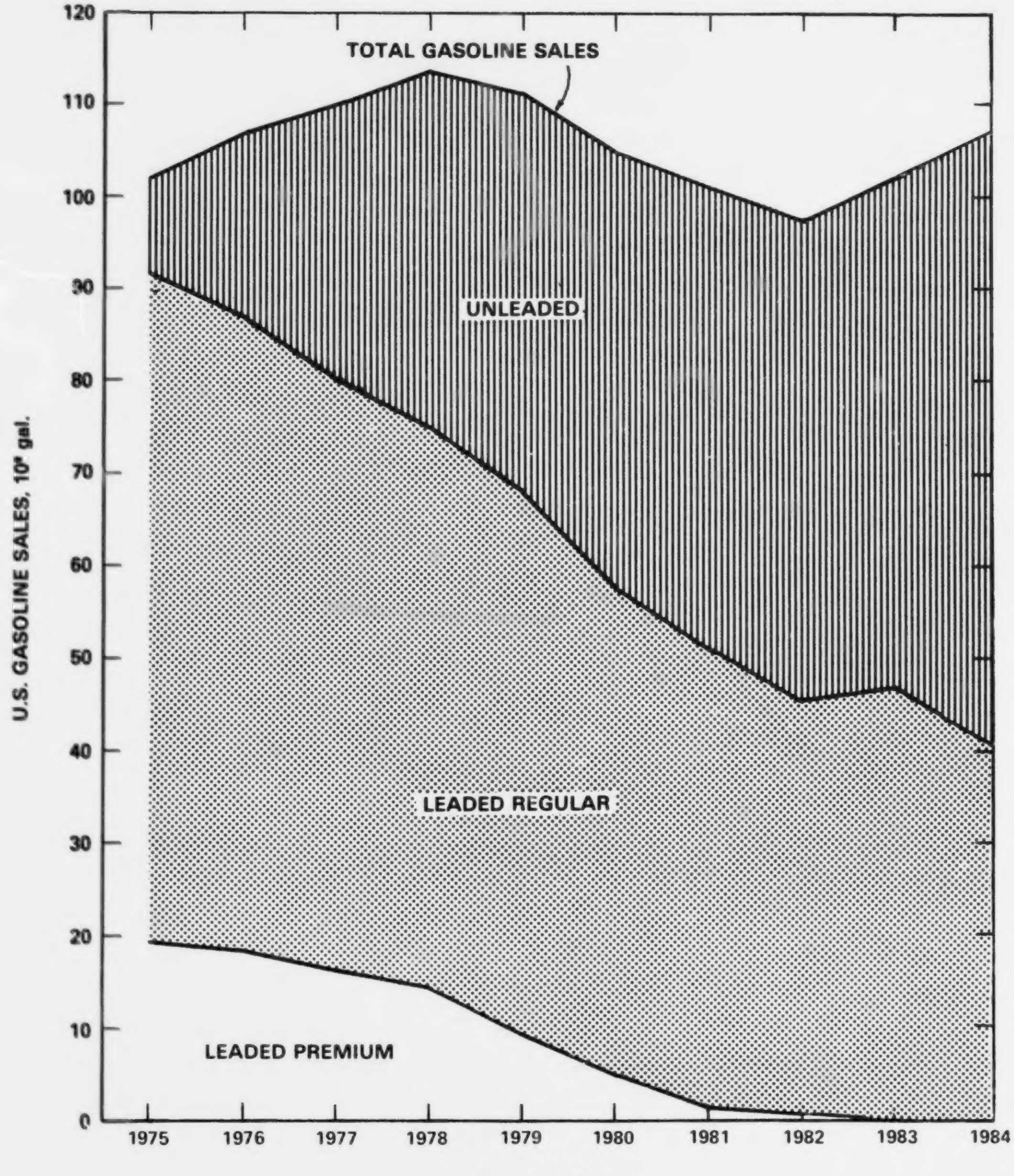

Figure 5-6. Trend in U.S. gasoline sales, 1975-1984.

Source: U.S. EPA (1985).

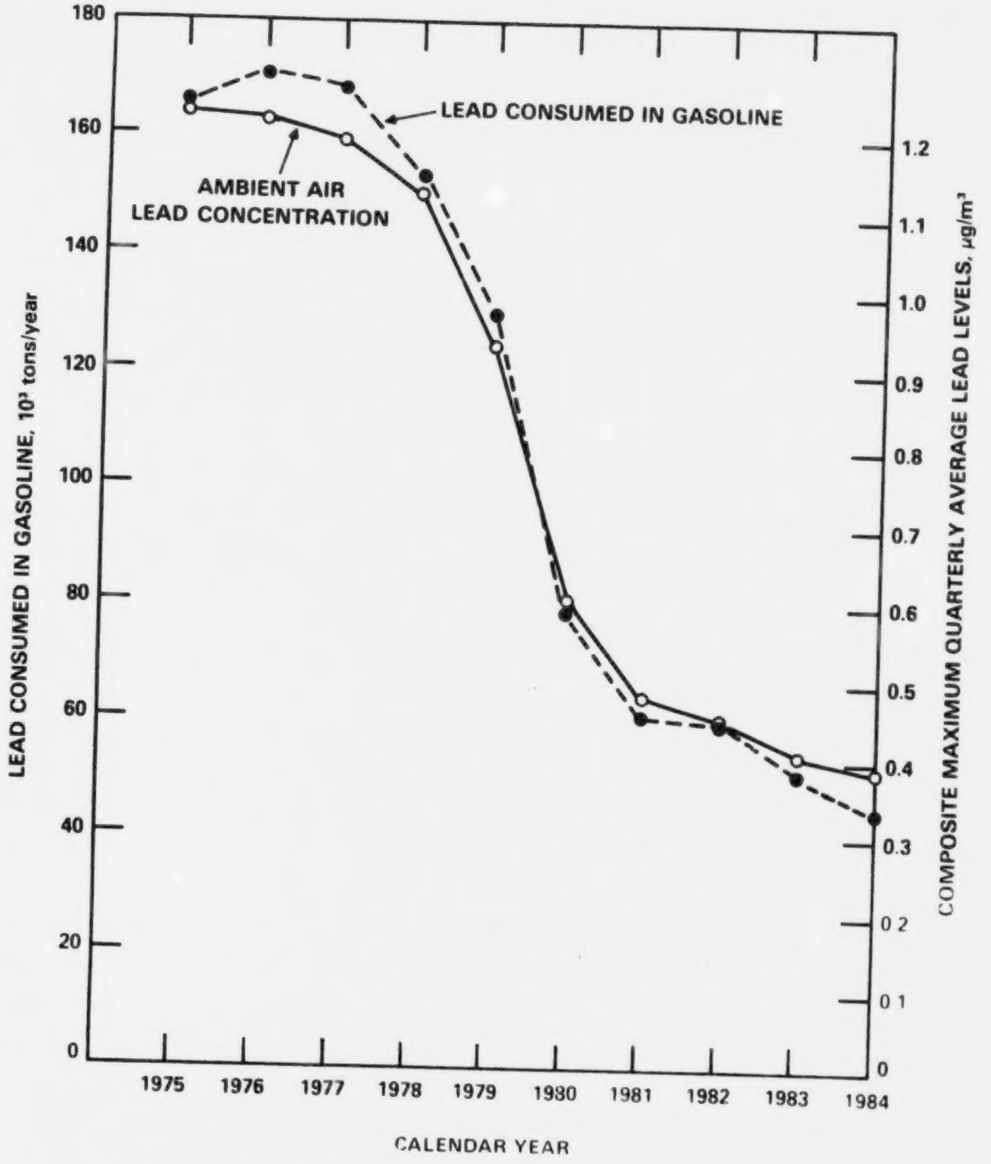

Figure 5-7. Lead consumed in gasoline and ambient lead concentrations, 1975-1984.

Source: U.S. Environmental Protection Agency (1985, 1986).

In the United States, a new source for lead emissions emerged in the mid-1960s with the opening of the "Viburnum Trend" or "New Lead Belt" in southeastern Missouri. The presence of eight mines and three accompanying lead smelters in this area makes it the largest lead-producing district in the world and has moved the United States into first place among the world's lead-producing nations.

Although some contamination of soil and water occurs as a result of such mechanisms as leaching from mine and smelter wastes, quantitative estimates of the extent of this contamination are not available. Spillage of ore concentrates from open trucks and railroad cars, however, is known to contribute significantly to contamination along transportation routes. For example, along two routes used by ore trucks in southeastern Missouri, lead levels in leaf litter ranged from 2000 - 5000 µg/g at the roadway, declining to a fairly constant 100 - 200 µg/g beyond about 400 ft from the roadway (Wixson et al., 1977).

Another possible source of land or water contamination is the disposal of particulate lead collected by air pollution control systems. The potential impact on soil and water systems from the disposal of dusts collected by these control systems has not been quantified.

5.4 SUMMARY

There is no doubt that atmospheric lead has been a component of the human environment since the earliest written record of civilization. Atmospheric emissions are recorded in glacial ice strata and pond and lake sediments. The history of these global emissions seems closely tied to production of lead by industrially oriented civilizations.

Although there are conflicting reports of the amount of lead emitted from natural sources, even the more liberal estimate (25×10^3 t/year, Nriagu, 1979) is dwarfed by the global emissions from anthropogenic sources (450×10^3 t/year).

Production of lead in the United States has remained steady at about 1.2×10^6 t/year for the past decade. The gasoline additive share of this market has dropped from 18 to 6.5 percent during the period 1971 - 1984. The contribution of gasoline lead to total atmospheric emissions has remained high, at 89 percent, as emissions from stationary sources have decreased at the same pace as from mobile sources. The decrease in stationary source emissions is due primarily to control of stack emissions, whereas the decrease in mobile source emissions is a result of switchover to unleaded gasolines. The decreasing use of lead in gasoline is projected to continue through 1990.

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6. TRANSPORT AND TRANSFORMATION

6.1 INTRODUCTION

This chapter describes the transition from the emission of lead particles into the atmosphere to their ultimate deposition on environmental surfaces, i.e., vegetation, soil, household dust, or water. Lead emissions at the tailpipe are typically around 24,000 μg/m^3 (38 x 10^4 μg Pb/Kg gas x 0.0838 Kg gas/m^3 air x 0.75 tailpipe efficiency), while in city air, ambient lead values are usually between 0.1 and 10 μg/m^3 (Dzubay et al., 1979; Reiter et al., 1977; also see Section 7.2.1.1.1). These reduced concentrations are the result of dilution of effluent gas with clean air and the removal of particles by wet or dry deposition. Characteristically, lead concentrations are highest in confined areas close to sources and are progressively reduced by dilution or deposition in air masses more removed from sources.

At any particular location and time, the concentration of lead found in the atmosphere depends on the proximity to the source, the amount of lead emitted from sources, and the degree of mixing provided by the motion of the atmosphere. It is possible to describe quantitatively the physics of atmospheric mixing in a variety of ways and, with some limiting assumptions, to develop simulation models that predict atmospheric lead concentrations. These models are not sensitive to short-term variations in air motion over a period of weeks or months because these variations are suppressed by integration over long periods of time.

In highly confined areas such as parking garages or tunnels, atmospheric lead concentrations can be 10-1000 times greater than values measured near roadways or in urban areas. In turn, atmospheric lead concentrations are usually about 2½ times greater in the central city than in residential suburbs. Rural areas have even lower concentrations.

Because lead emissions in the United States have declined dramatically in the past few years, the older lead concentration data on which recent dispersion studies are based may seem irrelevant to existing conditions. Such studies do in fact illustrate principles of atmospheric dispersion and are valid when applied to existing concentrations of lead with appropriate corrections (see Section 7.2.1.1).

Transformations that may occur during dispersion are physical changes in particle size distribution, chemical changes from the organic to the inorganic phase, and chemical changes in the inorganic phase of lead particles. Particle size distribution stabilizes within a few hundred kilometers of the sources, although atmospheric concentration continues to decrease with distance. Concentrations of organolead compounds are relatively small (1-6 percent of total lead) except in special situations where gasoline is handled or where engines are started cold within confined areas. Ambient organolead concentrations decrease more rapidly than inorganic lead, suggesting conversion from the organic to the inorganic phase during transport. Inorganic lead appears to convert from lead halides and oxides to lead sulfates.

Lead is removed from the atmosphere by wet or dry deposition. The mechanisms of dry deposition have been incorporated into models that estimate the flux of atmospheric lead to the earth's surface. Of particular interest is deposition on vegetation surfaces, since this lead may be incorporated into food chains. Between wet and dry deposition, it is possible to calculate an atmospheric lead budget that balances the emission inputs discussed in Section 5.3.3. with deposition outputs.

6.2 TRANSPORT OF LEAD IN AIR BY DISPERSION

6.2.1 Fluid Mechanics of Dispersion

Particles in air streams are subject to the same principles of fluid mechanics as particles in flowing water (Friedlander, 1977). On this basis, the authors of several texts have described the mathematical arguments for the mixing of polluted air with clean air (Benarie, 1980; Dobbins, 1979; Pasquill, 1974). If the airflow is steady and free of turbulence, the rate of mixing is constant along a concentration gradient and is a function of particle size (Dobbins, 1979). If the steady flow of air is interrupted by obstacles near the ground, turbulent eddies or vortices may be formed. Diffusivity is no longer constant with particle size and concentration but may be influenced by windspeed, atmospheric stability, and the nature of the obstacle. By making generalizations of windspeed, stability, and surface roughness, it is possible to construct models using a variable transport factor called eddy diffusivity (K), in which K varies in each direction, including vertically. There is a family of K-theory models that describe the dispersion of particulate pollutants.

The simplest K-theory model, which assumes that the surface is uniform and the wind is steady (Pasquill, 1974), produces a Gaussian plume, where the concentration of the pollutant decreases according to a normal or Gaussian distribution in both the vertical and horizontal directions. Although these models are the basis for most of the air quality simulations performed to date (Benarie, 1980), the assumptions of steady windspeed and smooth surface limit their use.

Some theoretical approaches, circumventing the constraints of the Gaussian models, have been adapted for studying long range transport (LRT) (more than 100 km) of pollutants. Johnson (1981) discusses 35 LRT models developed during the 1970s to describe the dispersion of atmospheric sulfur compounds. One family of models is based on the conservative volume element approach, where volumes of air are seen as discrete parcels having conservative meteorological properties, such as water vapor mixing ratio, potential temperature, and absolute vorticity (Benarie, 1980). The effect of pollutants on these parcels is expressed as a mixing ratio. These parcels of air may be considered to move along a trajectory that follows the advective wind direction. These models are particularly suitable for dealing with surface roughness, but they tend to introduce artifact diffusion or pseudodiffusion, which must be suppressed by calculation (Egan and Mahoney, 1972; Liu and Seinfeld, 1975; Long and Pepper, 1976).

An approach useful for estimating dispersion from a roadway derives from the similarity approach of Prandtl and Tietjens (1934). A mixing length parameter is related to the distance traveled by turbulent eddies during which violent exchange of material occurs. This mixing length is mathematically related to the square root of the shear stress between the atmosphere and the surface. Richardson and Proctor (1926) formulated these concepts in a law of atmospheric diffusion which was further extended to boundary layer concepts by Obukhov (1941). At the boundary layer, the turbulent eddy grows and its energy decreases with distance away from the source.

Although physical descriptions of turbulent diffusion exist for idealized circumstances such as isolated roadways and flat terrain, the complex flow and turbulence patterns of cities have defied theoretical description. The permeability of street patterns and turbulent eddy development in street canyons are two major problem areas that make modeling urban atmospheres difficult. Kotake and Sano (1981) have developed a simulation model for describing air flow and pollutant dispersion in various combinations of streets and buildings on two scales. A small scale, 2-20 m, is used to define the boundary conditions for 2-4 buildings and associated roadways. These subprograms are combined on a large scale of 50-500 meters. Simulations for oxides of nitrogen show nonlinear turbulent diffusion, as would be expected. The primary utility of this program is to establish the limits of uncertainty, the first step toward making firm predictions. It is likely that the development of more complete models of dispersion in complex terrains will become a reality in the near future.

None of the models described above have been tested for lead. The reason for this is simple. All of the models require sampling periods of 2 hours or less in order for the sample to conform to a well-defined set of meteorological conditions. In most cases, such a sample would be below the detection limits for lead. The common pollutant used to test models is SO_2, which can be measured over very short, nearly instantaneous, time periods. The question of whether gaseous SO_2 can be used as a surrogate for particulate lead in these models remains to be answered.

6.2.2 Influence of Dispersion on Ambient Lead Concentrations

Dispersion within confined situations, such as parking garages, residential garages and tunnels, and away from expressways and other roadways not influenced by complex terrain features depends on emission rates and the volume of clean air available for mixing. These factors are relatively easy to estimate and some effort has been made to describe ambient lead concentrations that can result under selected conditions. On an urban scale, the routes of transport are not clearly defined, but can be inferred from an isopleth diagram, i.e., a plot connecting points of identical ambient concentrations. These plots always show that lead concentrations are maximum where traffic density is highest.

Dispersion beyond cities to regional and remote locations is complicated by the facts that there are no monitoring network data from which to construct isopleth diagrams, that removal by deposition plays a more important role with time and distance, and that emissions from many different sources converge. Some techniques of source reconciliation are described, but these become less precise with increasing distance from major sources of lead. Dispersion from point sources such as smelters and refineries results in a concentration distribution pattern similar to urban dispersion, although the available data are notably less abundant.

6.2.2.1 Confined and Roadway Situations. Ingalls and Garbe (1982) used a variety of box and Gaussian plume models to calculate typical levels of automotive air pollutants that might be present in microscale (within 100 m of the source) situations with limited ventilation, such as garages, tunnels, and street canyons. Table 6-1 shows a comparison of six exposure situations, recomputed for a flat-average lead emission factor of 6.3 mg/km for roadway situations and 1.0 mg/min for garage situations. The roadway emission factor chosen corresponds roughly to values chosen by Dzubay et al. (1979) and Pierson and Brachaczek (1976) scaled to 1979 lead-use statistics. The parking garage factor was estimated from roadway factors by correction for fuel consumption (Ingalls and Garbe, 1982).

Confined situations, with low air volumes and little ventilation, allow automotive pollutant concentrations to reach one to three orders of magnitude higher than are found in open air. Thus, parking garages and tunnels are likely to have considerably higher ambient lead concentrations than are found in expressways with high traffic density or in city streets. Purdue et al. (1973) found total lead levels of 1.4-2.3 $\mu g/m^3$ in five of six U.S. cities in 1972. In similar samples from an underground parking garage, total lead was 11-12 $\mu g/m^3$. Vaitkus et al. (1974) developed a model for the transport of automotive lead that predicted an exponential decrease in air lead concentrations with distance, up to 100 m downwind from the roadway. Dzubay et al. (1979) found lead concentrations of 4-20 $\mu g/m^3$ in air over Los Angeles freeways in 1976; at nearby sites off the freeways, concentrations of 0.3-4.7 $\mu g/m^3$ were measured.

TABLE 6-1. SUMMARY OF MICROSCALE CONCENTRATIONS

Situation	Air lead concentration ($\mu g/m^3$)
Residential garage (1 mg Pb/min)	
Typical (30 second idle time)	80
Severe (5 min idle time)	670
Parking garage (1 mg Pb/min)	
Typical	40
Severe	560
Roadway tunnel (6.3 mg Pb/km)	
Typical	11
Severe	29
Street canyon (sidewalk receptor) (6.3 mg Pb/km)	
Typical a) 800 vehicles/hr	0.4
b) 1,600 vehicles/hr	0.9
Severe a) 800 vehicles/hr	1.4
b) 1,600 vehicles/hr	2.8
On expressway (wind: 315 deg. rel., 1 m/sec) (6.3 mg Pb/km)	
Typical	2.4
Severe	10

Data are recalculated from Ingalls and Garbe (1982) using 1979 lead emission factors. They show that air lead concentrations in a garage or tunnel can be two or three orders of magnitude higher than on streets or expressways. Typical conditions refer to neutral atmospheric stability and average daily traffic volumes. Severe conditions refer to maximum hourly traffic volume with atmospheric inversion. Emission rates are given in parentheses.

Tiao and Hillmer (1978) and Ledolter and Tiao (1979) have analyzed 3 years (1974-1977) of ambient air lead data from one site on the San Diego Freeway in Los Angeles, California. Particulate lead concentrations were measured at five locations: in the median strip and at distances of 8 and 30-35 m from the road edge on both sides of the road. Average lead concentrations at the 35 meter point were two- to fourfold lower than at the 8 m location (Tiao and Hillmer, 1978). An empirical model involving traffic count and traffic speed, which are related to road emissions, required only windspeed as a predictor of dispersion conditions.

Witz et al. (1982) found that meteorological parameters in addition to windspeed, such as inversion frequency, inversion duration, and temperature, correlate well with ambient levels of lead. At a different site near the San Diego freeway in Los Angeles, monthly ambient particulate lead concentrations and meteorological variables were measured about 100 meters from the roadway through 1980. Multiple linear regression analysis showed that temperature at 6 AM, windspeed, wind direction, and a surface-based inversion factor were important variables in accurately predicting monthly average lead concentrations. In this data set, lead values for December were about fivefold higher than those measured in the May to September summer season, suggesting that seasonal variations in wind direction and the occurrence of surface-based inversions favor high winter lead values. Unusually high early morning temperatures and windspeed during the winter increased dispersion and reduced lead concentration.

In a study of a newly constructed freeway near Melbourne, Australia, Clift et al. (1983) found that lead concentrations in the top centimeter of soil one meter from the road edge increased in lead concentration from 60 µg/g in March 1974 to 1250 µg/g in November 1980. The traffic density in November 1980 was 37,000 vehicles per day and the typical pattern of lead concentrations decreasing exponentially with distance from the road edge had developed. At 4-6 meters from the road edge, lead concentrations decreased to constant values, although these values were significantly higher than the pre-1974 concentrations.

In Philadelphia, a recent study of dispersion away from a major highway showed the zone of influence may extend farther downwind than previously expected (Burton and Suggs, 1984). The Philadelphia Roadway study was designed to measure the vertical (15 m) and horizontal (175 m) dispersion of large and small particles (Figure 6-1). Horizontally, air concentrations decreased exponentially at 2 m height for fine, coarse, and total Pb according to the following equations:

Coarse Pb	$C = 0.187 - (0.029 \times \ln D)$
Fine Pb	$C = 0.715 - (0.106 \times \ln D)$
Total Pb	$C = 0.903 - (0.135 \times \ln D)$

where C is the concentration of lead in air (µg/m^3) at the downwind distance, D (m), measured from the edge of the road. The numerical coefficients are specific for this site, and were found to vary with windspeed and traffic density.

Vertical profiles showed decreasing lead concentrations with increasing height for coarse, fine, and total particulate lead at 5 and 25 m downwind, although the effect was less pronounced at 25 m. Bullin et al. (1985) found similar results in Houston, somewhat tempered by greater mixing due to the presence of tall buildings.

6.2.2.2 Dispersion of Lead on an Urban Scale. In cities, air pollutants, including lead, that are emitted from automobiles tend to be highest in concentration in high traffic areas. Most U.S. cities have a well-defined central business district (CBD) where lead concentrations are highest. To illustrate the dispersion of lead experienced in cities, two cases are presented below.

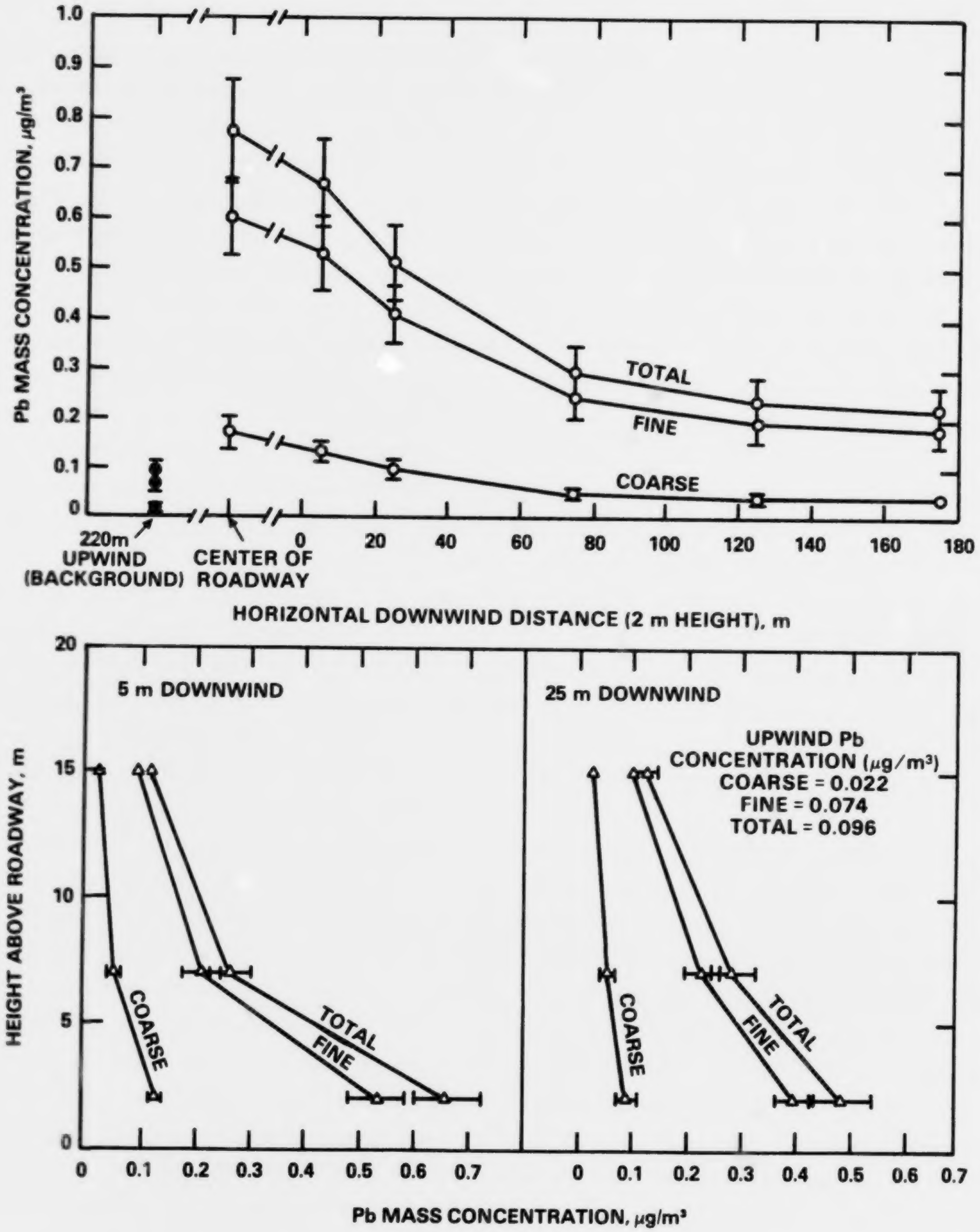


Figure 6-1. Vertical and horizontal distribution of lead downwind from a roadway in Philadelphia, PA.

Source: Burton and Suggs (1984).

For the South Coast Basin of Southern California, the area of high traffic density is more widespread than is characteristic of many cities. Ambient concentrations of lead tend to be more uniform. For example, Figures 6-2 and 6-3 show the average daily traffic by grid square and the contour plots of annual average lead concentration, respectively, for 1969 (Kawecki, 1978). In addition, Figure 6-3 shows annual average lead measured at nine sites in the basin for that year. It is clear that the central portion had atmospheric particulate lead concentrations of about 3 μg/m^3; the outer areas were in the range of 1-2 μg/m^3.

Reiter et al. (1977) have shown similar results for the town of Fort Collins, Colorado, for a 5.5-hr period in May of 1973. In that study, modeling results showed maximum lead concentrations in the center of town around 0.25 μg/m^3, which decreased to 0.1 μg/m^3 in the outermost region. Presumably, still lower values would be found at more remote locations.

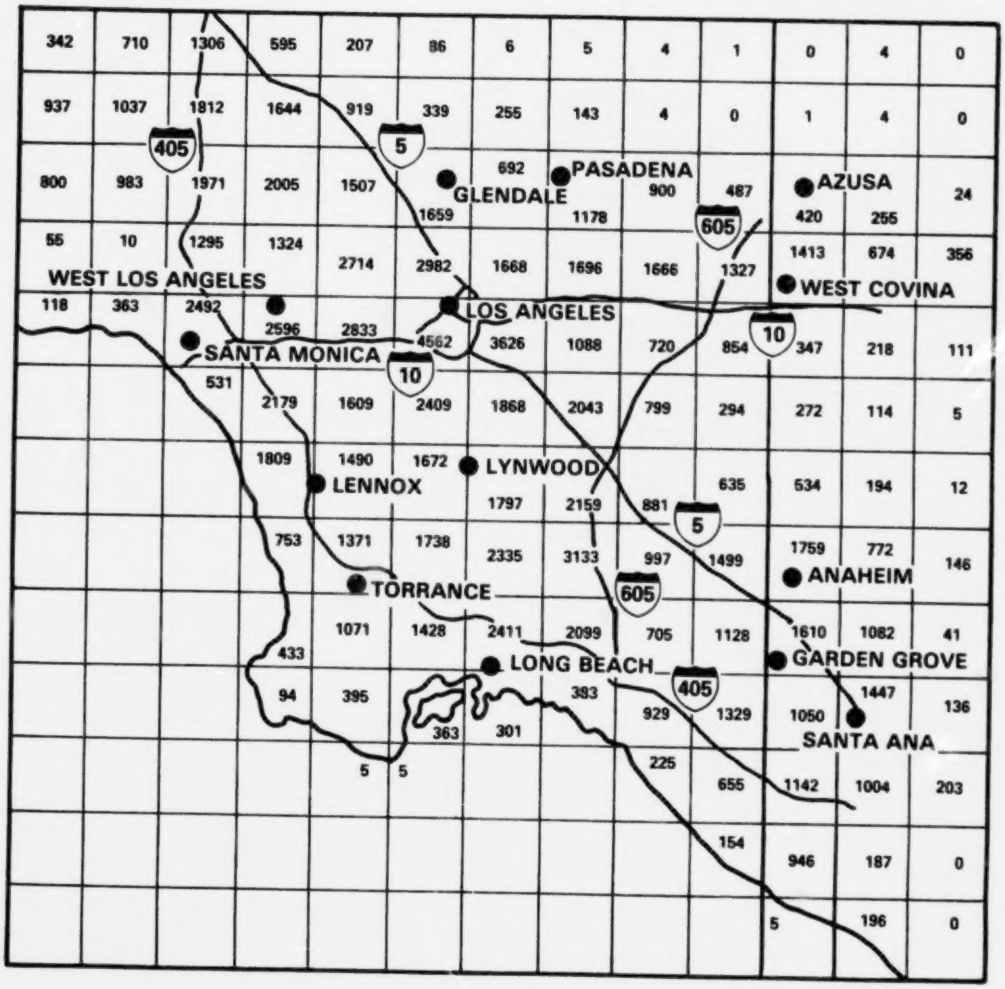
Apparently, then, lead in the air decreases 2- to 3-fold from maximum values in center city areas to well populated suburbs, with a further 2-fold decrease in the outlying areas. These modeling estimates are generally confirmed by measurement in the cases cited above and in the data presented in Section 7.2.1.

6.2.2.3 Dispersion from Smelter and Refinery Locations.

The 11 mines and 5 primary smelters and refineries shown in Figure 5-3 are not located in urban areas. Most of the 39 secondary smelters and refineries are likewise non-urban. Consequently, dispersion from these point sources should be considered separately, but in a manner similar to the treatment of urban regions. In addition to lead concentrations in air, concentrations in soil and on vegetation surfaces are often used to determine the extent of dispersion of plumes from smelters and refineries. In a study of smelters in Missouri, Dorn et al. (1976) found that 66 percent of the mass of lead was on particles smaller than 4.7 μm on a farm near a smelter (800 m from the smelter stack), whereas 73 percent were smaller than 4.7 μm on the control farm. These authors also noted seasonal differences in particle size distributions, with larger differences between the test and control farms during the winter than the spring or summer.

6.2.2.4 Dispersion to Regional and Remote Locations.

Beyond the immediate vicinity of urban areas and smelter sites, lead in air declines rapidly to concentrations of 0.1 to 0.5 μg/m^3. Two mechanisms responsible for this change are dilution with clean air and removal by deposition (Section 6.4). In the absence of monitoring networks that might identify the sources of lead in remote areas, two techniques of source identification have been used. Vector gradient analysis was attempted by Everett et al. (1979) and source reconciliation has been reported by Sievering et al. (1980) and Cass and McRae (1983). A third technique, isotopic composition, has been used to identify anthropogenic lead in air, sediments, soils, plants, and animals in urban, rural, and remote locations (Chow et al., 1975). Whereas this technique can often identify the source of lead, it has not yet been used to determine the mechanism of transport.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405
5
800
983
1971
2005
1507
GLENDALE
692
PASADENA
900
487
AZUSA
24
1659
1178
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WEST LOS ANGELES
WEST COVINA
118
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SANTA MONICA
4562
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LENNOX
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881
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753
1371
1738
2335
3133
997
1499
1759
772
146
ANAHEIM
TORRANCE
605
1071
1428
2411
2099
705
1128
1610
1082
41
433
LONG BEACH
GARDEN GROVE
405
94
395
383
1447
136
929
1329
1050
363
301
SANTA ANA
225
5
5
655
1142
1004
203
154
946
187
0
5
196
0

Figure 6-2. Spatial distribution of surface street and freeway traffic in the Los Angeles Basin (10^3 vehicle miles traveled/day) for 1979.

Source: Kawecki (1978).

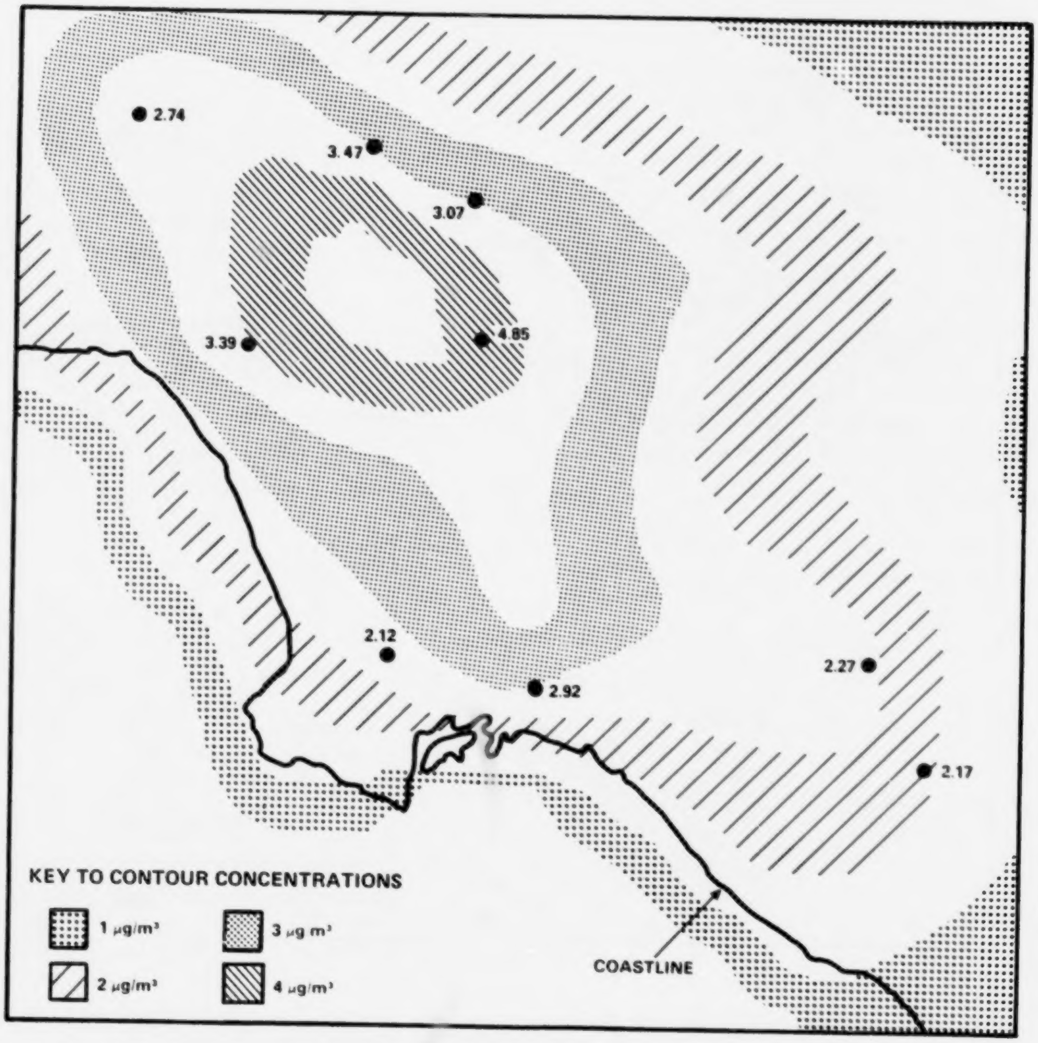
2.74
3.47
3.07
3.39
4.85
2.12
2.92
2.27
2.17
KEY TO CONTOUR CONCENTRATIONS
1 μg/m³
2 μg/m³
3 μg/m³
4 μg/m³
COASTLINE

Figure 6-3. Annual average suspended lead concentrations for 1969 in the Los Angeles Basin, calculated from the model of Cass (1975). The white zones between the patterned areas are transitional zones between the indicated concentrations.

Source: Kawecki (1978).

In vector gradient analysis, the sampler is oriented to the direction of the incoming wind vector, and samples are taken only during the time the wind is within a 30° arc of that vector. Other meteorological data are taken continuously. As the wind vector changes, a different sampler is turned on. A 360° plot of concentration vs. wind direction gives the direction from which the pollutant arrives at that location. Only one report of the use of this technique for lead occurs in the literature (Everett et al., 1979), and analysis of this experiment was complicated by the fact that in more than half the samples, the lead concentrations were below the detection limit. The study was conducted at Argonne National Laboratory and the results reflected the influence of automobile traffic east and northeast of this location.

Source reconciliation is based on the concept that each type of natural or anthropogenic emission has a unique combination of elemental concentrations. Measurements of ambient air, properly weighted during multivariate regression analysis, should reflect the relative amount of pollutant derived from each of several sources (Stolzenburg et al., 1982). Sievering et al. (1980) used the method of Stolzenberg et al. (1982) to analyze the transport of urban air from Chicago over Lake Michigan. They found that 95 percent of the lead in Lake Michigan air could be attributed to various anthropogenic sources, namely auto emissions, coal fly ash, cement manufacture, iron and steel manufacture, agricultural soil dust, construction soil dust, and incineration emissions. This information alone does not describe transport processes, but the study was repeated for several locations to show the changing influence of each source.

Cass and McRae (1983) used source reconciliation in the Los Angeles Basin to interpret 1976 NFAN data (see Sections 4.2.1 and 7.2.1.1) based on emission profiles from several sources. They developed a chemical element balance model, a chemical tracer model, and a multivariate statistical model. The chemical element balance model showed that 20 to 22 percent of the total suspended particle mass could be attributed to highway sources. The chemical tracer model permitted the lead concentration alone to represent the highway profile, since lead comprised about 12 percent of the mass of the highway generated aerosol. The multivariate statistical model used only air quality data without source emission profiles to estimate stoichiometric coefficients of the model equation. The study showed that single element concentrations can be used to predict the mass of total suspended particles.

Pacyna et al. (1985) used a receptor-oriented Lagrangian model to predict air concentrations in Spitsbergen, Norway, based on estimated emissions from the U.S.S.R. Compared to measured concentrations, the model was accurate for some metals, but overestimated the air concentration of lead by an average factor of 1.8. The consistent pattern in the ratio of estimated to measured air concentration led the authors to suggest that a more accurate estimate of lead emissions might correct the discrepancy.

A type of source reconciliation, chemical mass balance, has been used for many years by geochemists in determining the anthropogenic influence on the global distribution of elements. Two studies that have applied this technique to the transport of lead to remote areas are Murozumi et al. (1969) and Shirahata et al. (1980). In these studies, the influence of natural or crustal lead was determined by mass balance, and the relative influence of anthropogenic lead was established. In the Shirahata et al. (1980) study, the influence of anthropogenic lead was confirmed quantitatively by analysis of isotopic compositions in the manner of Chow et al. (1975).

Harrison and Williams (1982) determined air concentrations, particle size distributions, and total deposition flux at one urban and two rural sites in England. The urban site, which had no apparent industrial, commercial, or municipal emission sources, had an air-lead concentration of 3.8 $\mu g/m^3$, whereas the two rural sites were about 0.15 $\mu g/m^3$. The average particle size became smaller toward the rural sites, as the mass median aerodynamic diameter (MMAD) shifted downward from 0.5 µm to 0.1 µm. The total deposition flux will be discussed in Section 6.4.2.

Knowledge of lead concentrations in the oceans and glaciers provides some insight into the degrees of atmospheric mixing and long range transport. Tatsumoto and Patterson (1963), Chow and Patterson (1966), and Schaule and Patterson (1980) measured dissolved lead concentrations in sea water in the Mediterranean, in the Central North Atlantic (near Bermuda), and in the northeast Pacific, respectively. The profile obtained by Schaule and Patterson (1980) is shown in Figure 6-4. Surface concentrations in the Pacific (14 ng/kg) were found to be higher than those of the Mediterranean or the Atlantic, decreasing abruptly with depth to a relatively constant level of 1-2 ng/kg. The vertical gradient was found to be much less in the Atlantic (Figure 6-5). Tatsumoto and Patterson (1963) had earlier estimated an average surface lead concentration of 200 ng/kg in the northern hemispheric oceans. Chow and Patterson (1966) revised this estimate downward to 70 ng/kg. Below the mixing layer, there appears to be no difference between lead concentrations in the Atlantic and Pacific. These studies are significant in that they show that seawater concentrations during prehistoric times (below the mixing layer) were constant and much lower than modern seawater concentrations at the surface. From these data, it is possible to calculate present and prehistoric atmospheric inputs to the oceans (Schaule and Patterson, 1980), and by inference, the prehistoric concentrations of lead in air. They estimated the present inputs are 60-68 $ng/cm^2 \cdot yr$, which is 10-20 times the prehistoric rate.

Wiersma and Davidson (1985) have reviewed published data on trace metal concentrations (including lead) in the atmosphere at remote northern and southern hemispheric sites. The natural sources for such atmospheric trace metals include the oceans and the weathering of the

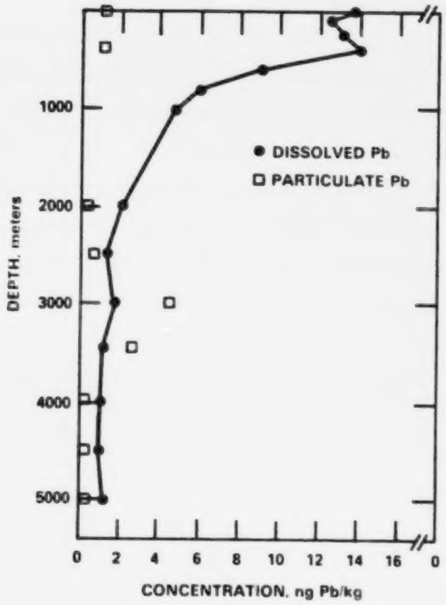

Figure 6-4. Profile of lead concentrations in the central northeast Pacific. Values below 1000 m are an order of magnitude lower than reported by Tatsumoto and Patterson (1963) and Chow and Patterson (1966).

Source: Schaule and Patterson (1980).

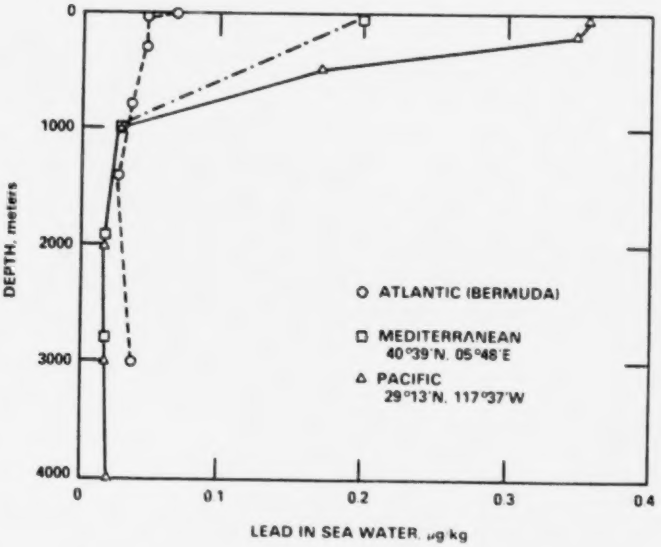

Figure 6-5. Lead concentration profiles in oceans show extensive contamination above the mixing layer (~ 1000 m).

Source: Chow and Patterson (1966).

earth's crust, while the anthropogenic source is particulate air pollution. Enrichment factors for concentrations relative to standard values for the oceans and the crust were calculated (Table 6-2); the crustal enrichment factors for the northern and southern hemispheres suggest that 90 percent of the particulate pollutants in the global troposphere are injected in the northern hemisphere. Since the residence times for particles in the troposphere are much less than the interhemispheric mixing time (Poet et al., 1972), it is unlikely that significant amounts of particulate pollutants can migrate from the northern to the southern hemisphere via the troposphere; however, this does not rule out stratospheric transfer.

TABLE 6-2. ENRICHMENT OF ATMOSPHERIC AEROSOLS OVER CRUSTAL ABUNDANCE ($EF_{crustal}$) IN REMOTE AREAS OF THE NORTHERN AND SOUTHERN HEMISPHERES

Element	Remote air concentration range[a], ng/m^3	Global $EF_{crustal}$	Remote continental $EF_{crustal}$	Remote marine $EF_{crustal}$
Al	0.3-1200	1.0	1.0	1.0
Si	21-3900	0.84	0.7	0.7
Fe	0.25-660	1.3	1.5(N)[b] 1.0(S)	2.5(N) 1.0(S)
Mn	0.0067-190	1.5	2.0(N) 1.0(S)	3.0(N) 1.0(S)
Ca	1.9-1600	1.8	1.5	8.0
Co	0.0017-1.0	1.9	1.5(N) 0.9(S)	4.0(N) 0.9(S)
V	0.001-1.5	3.3	1.5	15(N) 1.5(S)
Cr	0.01-7.0	3.6	6.0(N) 1.0(S)	20(N) 1.0(S)
Cu	0.06-110	25	20	150
Zn	0.035-110	50	80	400
Sb	0.002-0.9	211	500	2000
Pb	0.027-97	320	2000(N) 80(S)	2000(N) 150(S)
Cd	0.02-2.2	1100	2000	5000
Se	0.006-1.4	3500	1000	6000

[a] See text for explanation of the relationship between air concentration and $EF_{crustal}$.

[b] (N) = northern hemisphere; (S) = southern hemisphere.

Source: data from Wiersma and Davidson (1985).

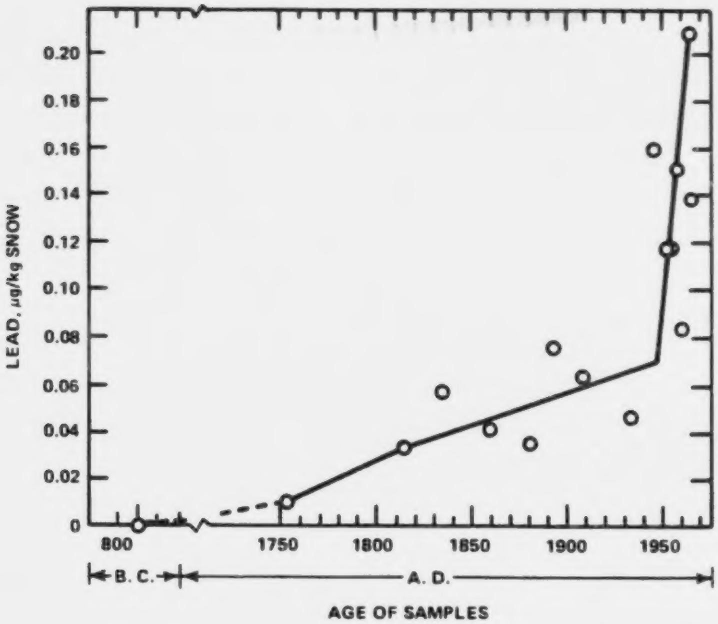

Figure 6-6. Lead concentration profile in snow strata of Northern Greenland.

Source: Murozumi et al. (1969).

Murozumi et al. (1969) have shown that long range transport of lead particles emitted from automobiles has significantly polluted the polar glaciers. They collected samples of snow and ice from Greenland and the Antarctic. As shown in Figure 6-6, they found that the concentration of lead in Greenland varied inversely with the geological age of the sample. The authors attribute the gradient increase after 1750 to the Industrial Revolution and the accelerated increase after 1940 to the increased use of lead alkyls in gasoline. The most recent levels found in the Antarctic snows (not shown on Figure 6-6) were less than those found in Greenland by a factor of 10 or more. Before 1940, the concentrations in the Antarctic were below the detectable level (<0.001 μg/kg) and have risen to 0.2 μg/kg in recent snow.

Evidence from remote areas of the world suggests that lead and other fine particle components are transported substantial distances, up to thousands of kilometers, by general weather systems. The degree of surface contamination of remote areas with lead depends both on weather influences and on the degree of air contamination. However, even in remote areas, man's primitive activities can play an important role in atmospheric lead levels. Davidson et al. (1982) have shown that there are significant levels of fine particle lead, up to 0.5 μg/m^3, in remote villages in Nepal. The apparent source is combustion of dried yak dung, which contains small amounts of naturally occurring lead derived from plant life in those remote valleys.

6.3 TRANSFORMATION OF LEAD IN AIR

6.3.1 Particle Size Distribution

Whitby et al. (1975) placed atmospheric particles into three different size regimes: the nuclei mode (<0.1 μm), the accumulation mode (0.1-2 μm) and the large particle mode (>2 μm). At the source, lead particles are generally in the nuclei and large particle modes. Large particles are removed by deposition close to the source and particles in the nuclei mode diffuse to surfaces or agglomerate while airborne to form larger particles of the accumulation mode. Thus it is in the accumulation mode that particles are dispersed great distances.

Pierson and Brachaczek (1983) reported particle size distributions for ambient air that were skewed farther to the right (more large particles) than in a roadway tunnel, where vehicle exhaust must be dominant (Figure 6-7). The large particles may have been deposited in the roadway itself and small particles may have agglomerated during transport away from the roadway (see Section 5.3.3.1). Since 40 to 1,000 μm particles are found in gutter debris, deposition of large particles appears confirmed (Pierson and Brachaczek, 1976, 1983).

Particle size distributions reported by Huntzicker et al. (1975) show bimodal distributions for on-roadway samples, with peak mass values at about 0.1 and 10 μm. For off-roadway Pasadena samples, there is no evidence of bimodality and only a broad maximum in lead mass between 0.1 and 1 μm.

In cities or in rural areas, there is a remarkable consistency in lead particle size range. For example, Robinson and Ludwig (1964) report cascade impactor MMAD values for lead ranging from 0.23 to 0.3 μm in six U.S. cities and three rural areas. Stevens et al. (1978) have reported dichotomous sampler data for six U.S. cities, as shown in Table 6-3, and Stevens et al. (1980, 1982) have reported similar results for remote locations. Virtually every other study reported in the literature for Europe, South America, and Asia has come to the conclusion that ambient urban and rural air contains predominantly fine particles (Cholak et al., 1968; De Jonghe and Adams, 1980; Durando and Aragon, 1982; Lee et al., 1968; Htun and Ramachandran, 1977). The size distributions of lead-bearing particles in ambient air from several global locations are discussed further in Section 7.2.1.3.1 and shown in Figure 7-5.

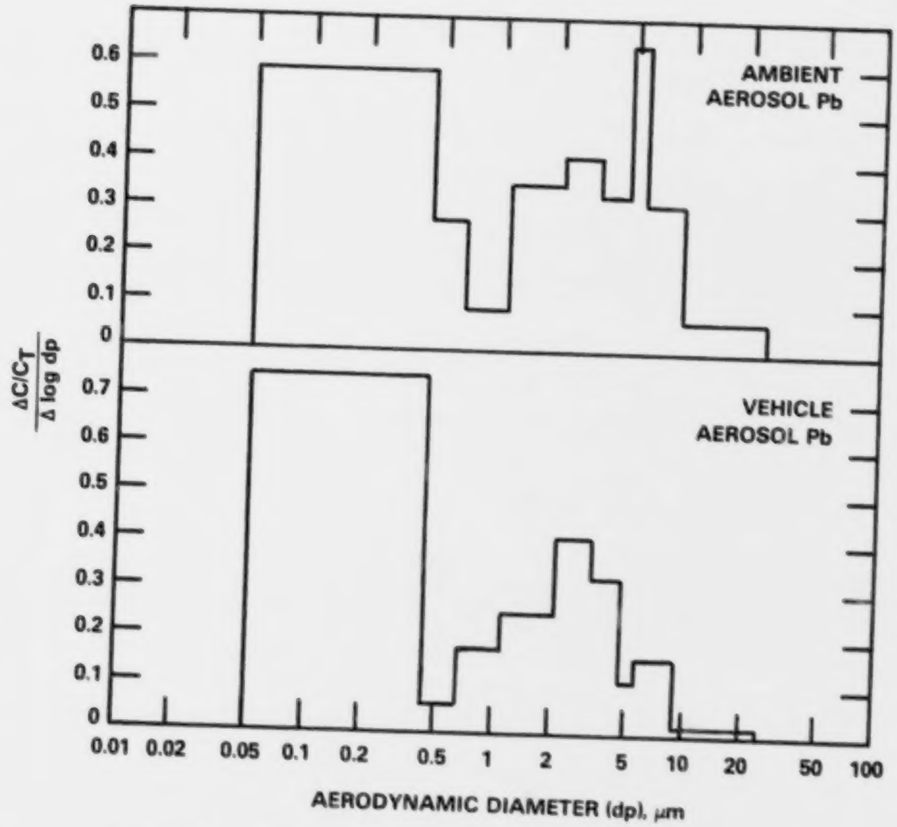

Figure 6-7. Typical airborne mass size distribution patterns for ambient and vehicle aerosol lead. ΔC represents the airborne lead concentrations in each size range. C_T is the total airborne lead concentration in all size ranges, and dp is the aerodynamic particle diameter.

Source: Data from Pierson and Brachaczek (1983).

TABLE 6-3. DISTRIBUTION OF LEAD IN TWO SIZE FRACTIONS AT SEVERAL SITES IN THE UNITED STATES

Location	Date	Fine[a]	Coarse[a]	F/C ratio
New York, NY	2/1977	1.1	0.18	6.0
Philadelphia, PA	2-3/1977	0.95	0.17	5.6
South Charleston, WV	4-8/1976	0.62	0.13	4.6
St. Louis, MO	12/1975	0.83	0.24	3.4
Portland, OR	12/1977	0.87	0.17	5.0
Glendora, CA	3/1977	0.61	0.09	6.7
Average				5.2

[a] Data are in μg/m^3.

Source: Stevens et al. (1978).

The data in Table 6-3 indicate that there is about five times more lead associated with small particles than large particles in urban atmospheres. It appears that lead particle size distributions are stabilized close to roadways and remain constant with transport into remote environments (Gillette and Winchester, 1972).

6.3.2 Organic (Vapor Phase) Lead in Air

Small amounts of lead additives may escape to the atmosphere by evaporation from fuel systems or storage facilities. Tetraethyllead (TEL) and tetramethyllead (TML) photochemically decompose when they reach the atmosphere (Huntzicker et al., 1975; National Air Pollution Control Administration, 1965). The lifetime of TML is longer than that of TEL. Laveskog (1971) found that transient peak concentrations of organolead up to 5,000 μg/m^3 in exhaust gas may be reached in a cold-started, fully choked, and poorly tuned vehicle. If a vehicle with such emissions were to pass a sampling station on a street where the lead level might typically be 0.02-0.04 μg/m^3, a peak of about 0.5 μg/m^3 could be measured as the car passed by. The data reported by Laveskog were obtained with a procedure that collected very small (100 ml), short-time (10 min) air samples. Harrison et al. (1975) found levels as high as 0.59 μg/m^3 (9.7 percent of total lead) at a busy gasoline service station in England. Grandjean and Nielsen (1979), using GC-MS techniques, found elevated levels (0.1 μg/m^3) of TML in city streets in Denmark and Norway. These authors attributed these results to the volatility of TML compared with TEL.

A number of studies have used gas absorbers behind filters to trap vapor-phase lead compounds (see Section 4.2.2.5). Because it is not clear that all the lead captured in the backup traps is, in fact, in the vapor phase in the atmosphere, "organic" or "vapor phase" lead is an operational definition in these studies. Purdue et al. (1973) measured both particulate and organic lead in atmospheric samples. They found that the vapor phase lead was about 5 percent of the total lead in most samples (see Section 5.3.3.1). The results are consistent with the studies by Huntzicker et al. (1975) who reported an organic component of 6 percent of the total airborne lead in Pasadena for a 3-day period in June, 1974, and by Skogerboe (1976), who measured fractions in the range of 4 to 12 percent at a site in Fort Collins, Colorado. It is noteworthy, however, that in an underground garage, total lead concentrations were approximately five times greater than those in ambient urban atmospheres, and the organic lead increased to approximately 17 percent.

Harrison et al. (1979) report typical organolead percentages in ambient urban air of 1-6 percent. Rohbock et al. (1980) reported higher fractions, up to 20 percent, but the data and interpretations have been questioned by Harrison and Laxen (1981). Rohbock et al. (1980) and De Jonghe and Adams (1980) report one to two orders of magnitude decrease in organolead concentrations from the central urban areas to residential areas. A review by Nielsen (1984) documents the concentrations of organolead in partially enclosed areas such as gas stations, parking garages, car repair shops, and tunnels, and in open urban and rural areas in the U.S. and Europe. Mean concentrations varied from 0.15 to 3.5 μg organolead/m^3 in enclosed areas and 0.014 to 0.47 in open urban areas.

6.3.3 Chemical Transformations of Inorganic Lead in Air

Lead is emitted into the air from automobiles as lead halides and as double salts with ammonium halides (e.g., $PbBrCl \cdot 2NH_4Cl$). From mines and smelters, $PbSO_4$, $PbO \cdot PbSO_4$, and PbS appear to be the dominant species. In the atmosphere, lead is present mainly as the sulfate with minor amounts of halides. It is not completely clear just how the chemical composition changes in transport.

Biggins and Harrison (1978, 1979) have studied the chemical composition of lead particles in exhaust and in city air in England by X-ray diffractometry. These authors reported that the dominant exhaust forms were PbBrCl, $PbBrCl \cdot 2NH_4Cl$, and $\alpha\text{-}2PbBrCl \cdot NH_4Cl$, in agreement with the earlier studies of Hirschler and Gilbert (1964) and Ter Haar and Bayard (1971).

At sampling sites in Lancaster, England, Biggins and Harrison (1978, 1979) found $PbSO_4 \cdot (NH_4)_2SO_4$, and $PbSO_4 \cdot (NH_4)_2BrCl$ together with minor amounts of the lead halides and double salts found in auto exhaust. These authors suggested that emitted lead halides react with acidic gases or aerosol components (SO_2 or H_2SO_4) on filters to form substantial levels of sulfate salts. It is not clear whether reactions with SO_4^{2-} occur in the atmosphere or on the sample filter.

The ratio of Br to Pb is often cited as an indication of automotive emissions. From the mixtures commonly used in gasoline additives, the mass Br/Pb ratio should be 0.4-0.5 (Pierson and Brachaczek, 1976, 1983; Dzubay et al., 1979; Dietzmann et al., 1981; Lang et al., 1981). However, several authors have reported loss of halide, preferentially bromine, from lead salts in atmospheric transport (Dzubay and Stevens, 1973; Pierrard, 1969; Ter Haar and Bayard, 1971). Both photochemical decomposition (Lee et al., 1971; Ter Haar and Bayard, 1971) and acidic gas displacement (Robbins and Snitz, 1972) have been postulated as mechanisms. Chang et al. (1977) have reported only very slow decomposition of lead bromochloride in natural sunlight; currently the acid displacement of halide seems to be the most likely mechanism. O'Connor et al. (1977) have compared roadside and suburban-rural aerosol samples from western Australia and reported no loss in bromine; low levels of SO_2 and sulfate aerosol could account for that result. Harrison and Sturges (1983) warn of several other factors that can alter the Br/Pb ratio. Bromine may pass through the filter as hydrogen bromide gas, lead may be retained in the exhaust system, or bromine may be added to the atmosphere from other sources, such as marine aerosols. They concluded that Br/Pb ratios are only crude estimates of automobile emissions, and that this ratio would decrease with distance from the highway from 0.39 to 0.35 at less proximate sites to 0.25 in suburban residential areas. For an aged aerosol, the Br/Pb mass ratio is usually about 0.22.

Habibi et al. (1970) studied the composition of auto exhaust particles as a function of particle size. Their main conclusions follow:

1. Chemical composition of emitted exhaust particles is related to particle size.
    a. Very large particles, greater than 200 µm, have a composition similar to lead-containing material deposited in the exhaust system, confirming that they have been emitted from the exhaust system. These particles contain approximately 60 to 65 percent lead salts, 30 to 35 percent ferric oxide (Fe_2O_3), and 2 to 3 percent soot and carbonaceous material. The major lead salt is lead bromochloride (PbBrCl), with (15 to 17 percent) lead oxide (PbO) occurring as the 2PbO·PbBrCl double salt. Lead sulfate and lead phosphate account for 5 to 6 percent of these deposits. (These compositions resulted from the combustion of low-sulfur and low-phosphorus fuel.)

    b. PbBrCl is the major lead salt in particles of 2 to 10 µm equivalent diameter, with $2PbBrCl \cdot NH_4Cl$ present as a minor constituent.

    c. Submicrometer-sized lead salts are primarily $2PbBrCl \cdot NH_4Cl$.

2. Lead-halogen molar ratios in particles of less than 10 μm MMAD indicate that much more halogen is associated with these solids than the amount expected from the presence of $2PbBrCl \cdot NH_4Cl$, as identified by X-ray diffraction. This is particularly true for particles in the 0.5 to 2 μm size range.

3. There is considerably more soot and carbonaceous material associated with fine-mode particles than with coarse mode particles re-entrained after having been deposited following emission from the exhaust system. This carbonaceous material accounts for 15 to 20 percent of the fine particles.

4. Particulate matter emitted under typical driving conditions is rich in carbonaceous material. There is substantially less material emitted under continuous hot operation.

5. Only small quantities of $2PbBrCl \cdot NH_4Cl$ were found in samples collected at the tailpipe from the hot exhaust gas. Its formation therefore takes place primarily during cooling and mixing of exhaust with ambient air.

Foster and Lott (1980) used X-ray diffractometry to study the composition of lead compounds associated with ore handling, sintering, and blast furnace operations around a lead smelter in Missouri. Lead sulfide was the main constituent of those samples associated with ore handling and fugitive dust from open mounds of ore concentrate. The major constituents from sintering and blast furnace operations appeared to be $PbSO_4$ and $PbO \cdot PbSO_4$, respectively.

6.4 REMOVAL OF LEAD FROM THE ATMOSPHERE

Before atmospheric lead can have any effect on organisms or ecosystems, it must be transferred from the air to a surface by wet or dry deposition.

6.4.1 Dry Deposition

6.4.1.1 Mechanisms of Dry Deposition. The theory and mechanics of particle deposition from the atmosphere to smooth surfaces are fairly well understood (Friedlander, 1977). Transfer by dry deposition requires that the particle move from the main airstream through the boundary layer to a surface. The boundary layer is defined as the region of minimal air flow immediately adjacent to that surface. The thickness of the boundary layer depends mostly on the windspeed and roughness of the surface. Schack et al. (1985) have extended particle deposition theory to include completely rough surfaces, such as terrestrial surfaces.

Airborne particles do not follow a smooth, straight path in the airstream. On the contrary, the path of a particle may be affected by micro-turbulent air currents, gravitation, or inertia. There are several mechanisms that may alter the particle path enough to cause transfer to a surface. These mechanisms are a function of particle size, windspeed, and surface characteristics.

Particles larger than a few micrometers in diameter are influenced primarily by sedimentation, where the particle accelerates downward until aerodynamic drag is exactly balanced by gravitational force. The particle continues at this velocity until it reaches a surface. Sedimentation is not influenced by horizontal windspeed or surface characteristics. Particles moving in an airstream may be removed by impaction whenever they are unable to follow the airstream around roughness elements of the surface, such as leaves, branches, or tree trunks. In this case, the particle moves parallel to the airstream and strikes a surface perpendicular to the airstream. A related mechanism, turbulent inertial deposition, occurs when a particle encounters turbulence within the airstream causing the particle to move perpendicular to the airstream. It may then strike a surface parallel to the airstream. In two mechanisms, wind eddy diffusion and interception, the particle remains in the airstream until it is transferred to a surface. With wind eddy diffusion, the particle is transported downward by turbulent eddies. Interception occurs when the particle in the airstream passes within one particle radius of a surface. This mechanism is more a function of particle size than windspeed. The final mechanism, Brownian diffusion, is important for very small particles at very low windspeeds. Brownian diffusion is motion, caused by random collision with molecules, in the direction of a decreasing concentration gradient.

Transfer from the main airstream to the boundary layer is usually by sedimentation or wind eddy diffusion. From the boundary layer to the surface, transfer may be by any of the six mechanisms, although those that are independent of windspeed (sedimentation, interception, Brownian diffusion) are more likely. Determining deposition onto rough surfaces requires information of the height, shape and density of protrusions from the surface into the boundary layer (Schack et al., 1985). If dry deposition is seen as a two-step process, diffusion through the boundary layer and interception by the surface, then for rough surfaces with rapid eddy diffusion, interception by the protrusion surfaces becomes the rate-limiting step. Consequently, surfaces such as water, grass, or bare rocks can be evaluated using a general correlation with a reference surface (Schack et al., 1985), and a more complete understanding of dry deposition to natural surfaces may be possible with the application of these experimental results.

6.4.1.2 Dry deposition models. A particle influenced only by sedimentation may be considered to be moving downward at a specific velocity usually expressed in cm/sec. Similarly, particles transported to a surface by any mechanism are said to have an effective deposition velocity (V_d), which is an expression of the rate of particle mass transfer measured by accumulation on a surface as a function of time and air concentration. This relationship is expressed in the equation:

$$V_d = J/C \tag{6-1}$$

where J is the flux or accumulation expressed in $ng/cm^2 \cdot s$ and C is the air concentration in ng/cm^3. The units of V_d become cm/sec.

Several recent models of dry deposition have evolved from the theoretical discussion of Fuchs (1964) and the wind tunnel experiments of Chamberlain (1966). From those early works, it was obvious that the transfer of particles from the atmosphere to the Earth's surface involved more than rain or snow. The models of Slinn (1982) and Davidson et al. (1982) are particularly useful for lead deposition and were strongly influenced by the theoretical discussions of fluid dynamics by Friedlander (1977). Slinn's model considers a multitude of vegetation parameters to find several approximate solutions for particles in the size range of 0.1-1.0 µm. In the absence of appropriate field studies, Slinn (1982) estimates deposition velocities of 0.01-0.1 cm/sec.

The model of Davidson et al. (1982) is based on detailed vegetation measurements and wind data to predict a V_d of 0.05-1.0 cm/sec. Deposition velocities are specific for each vegetation type. This approach has the advantage of using vegetation parameters of the type made for vegetation analysis in ecological studies (density, leaf area index (LAI), height, diameter) and thus may be applicable to a broad range of vegetation types for which data are already available in the ecological literature.

Both models show a decrease in deposition velocity with decreasing particle size down to about 0.1-0.2 µm, followed by an increase in V_d with decreasing diameter from 0.1 to 0.001 cm/sec. On a log plot of V_d versus diameter, this curve is v-shaped (Sehmel, 1980), and the plots of several vegetation types show large changes (10X) in minimum V_d, although the minima commonly occur at about the same particle diameter (Figure 6-8). Although shown on the diagram, particles larger than 0.1 µm diameter are not likely to have a density as great as 11.5 g/km^3.

In summary, it is not correct to assume that air concentration and particle size alone determine the flux of lead from the atmosphere to terrestrial surfaces. The type of vegetation canopy and the influence of the canopy on windspeed are important predictors of dry deposition. Both of these models predict deposition velocities more than one order of magnitude lower than reported in several earlier studies (e.g., Sehmel and Hodgson, 1976).

6.4.1.3 Calculation of Dry Deposition. The data required for calculating the flux of lead from the atmosphere by dry deposition are leaf area index (LAI), windspeed, deposition velocity, and air concentration by particle size. The LAI should be total surface rather than upfacing surface, as used in photosynthetic productivity measurements. LAI's should also be expressed for the entire community rather than by individual plant, in order to incorporate

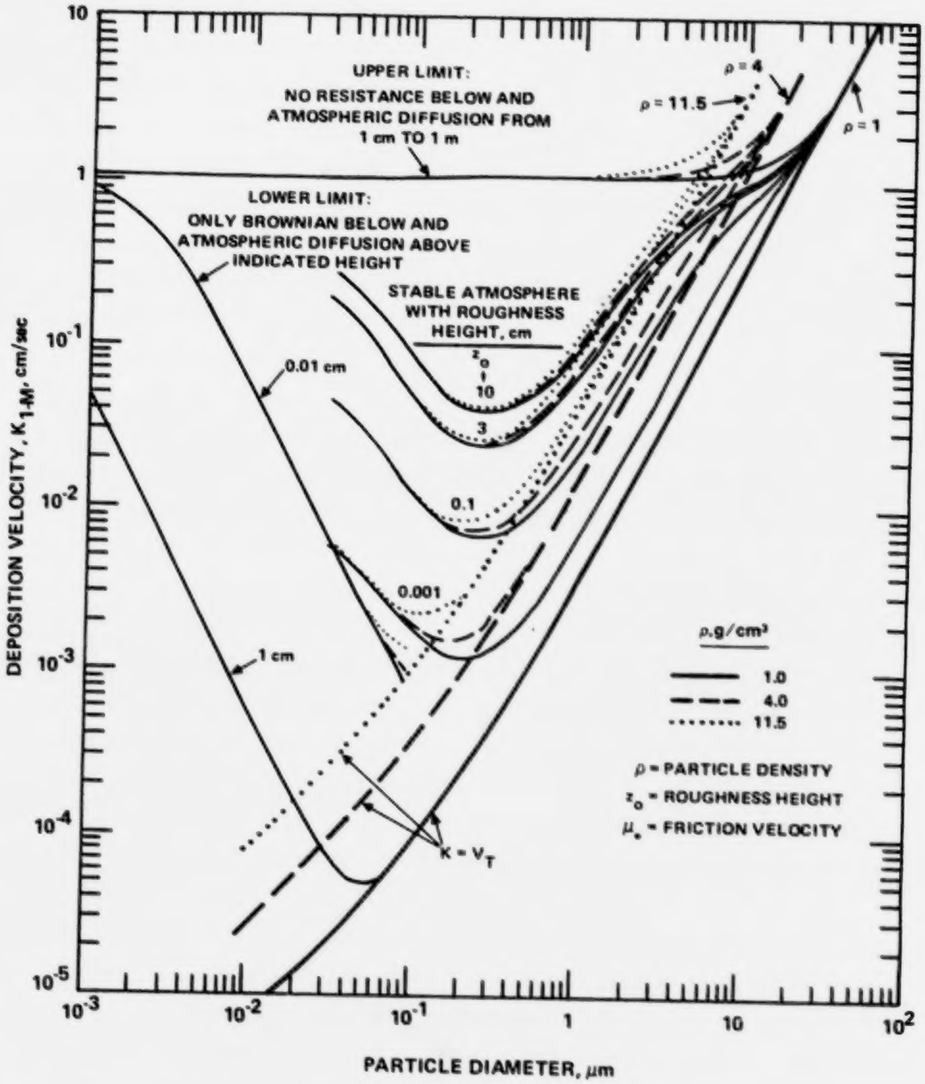

Figure 6-8. Predicted deposition velocities at 1 m for $\mu_* = 30$ cm s^{-1} and particle densities of 1, 4, and 11.5 cm^{-3}.

Source: Sehmel (1980).

variations in density. Some models use a more generalized surface roughness parameter, in which case the deposition velocity may also be different.

The value selected for V_d depends on the type of vegetation, usually described as either short (grasses or shrubs) or tall (forests). For particles with an MMAD of about 0.5 µm, Hicks (1979) gives values for tall vegetation deposition velocity from 0.1-0.4 cm/sec. Lannefors et al. (1983) estimated values of 0.2-0.5 cm/sec in the particle size range of 0.06-2.0 µm in a coniferous forest. For lead, with an MMAD of 0.55 µm, they measured a deposition velocity of 0.41 cm/sec. In a series of articles (Wiman and Agren, 1985; Wiman and Lannefors, 1985; and Wiman et al., 1985), this research group has described the modeling parameters required to define deposition in coniferous forests. They found a significant depletion of aerosols from the forest edge to the interior, and distinct edge effects, for larger particles, but nearly negligible depletion and edge effects for submicron particles. This suggests that because lead is borne primarily on particles less than 1 µm, lead deposition within a forest may be comparable to open grasslands and other vegetation types.

6.4.1.4 Field Measurements of Dry Deposition on Surrogate and Natural Surfaces. Several investigators have used surrogate surface devices similar to those described in Section 4.2.2.4. These data are summarized in Table 6-4. The few studies available on deposition to vegetation surfaces show deposition rates comparable to those of surrogate surfaces and deposition velocities in the range predicted by the models discussed above. A study to compare vegetation washing and several types of surrogate surfaces was reported by Dolske and Gatz (1984). Although the study emphasizes sulfate particles, the devices and techniques are similar to those used for lead. One important observation was that surrogate surface devices may be more representative of actual deposition if the device has a very shallow rim or no rim at all. Therefore, the data of Table 6-4 do not include measurements made with deposition buckets. In Section 6.4.3, these data are used to show that global emissions are in approximate balance with global deposition. It is reasonable to expect that future refinements of field measurements and model calculations will permit more accurate estimates of dry deposition in specific regions or under specific environmental conditions.

6.4.2 Wet Deposition

Wet deposition includes removal by rainout and washout. Rainout occurs when particulate matter is present in the supersaturated environment of a growing cloud. The small particles (0.1 to 0.2 µm) act as nuclei for the formation of small droplets, which grow into raindrops (Junge, 1963). Droplets also collect particles under 0.1 µm by Brownian motion and by the water-vapor gradient. These processes are referred to as rainout. Washout, on the other hand, occurs when falling raindrops collect particles by diffusion and impaction on the way to

TABLE 6-4. SUMMARY OF SURROGATE AND VEGETATION SURFACE DEPOSITION OF LEAD

Depositional surface	Deposition, ng Pb/cm^2·day	Air conc, ng/m^3	Deposition velocity, cm/sec	Reference
Tree leaves (Paris)	0.38	---	0.086	1
Tree leaves (Tennessee)	0.29-1.2	---	---	2
Plastic disk (remote California)	0.02-0.08	13-31	0.05-0.4	3
Plastic plates (Tennessee)	0.29-1.5	110	0.05-0.06	4
Tree leaves (Tennessee)	---	110	0.005	4
Snow (Greenland)	0.004	0.1-0.2	0.1	5
Grass (Pennsylvania)	---	590	0.2-1.1	6
Coniferous forest (Sweden)	0.74	21	0.41	7

1. Servant, 1975.
2. Lindberg et al., 1982.
3. Elias and Davidson, 1980.
4. Lindberg and Harriss, 1981.
5. Davidson et al., 1981.
6. Davidson et al., 1982.
7. Lannefors et al., 1983.

the ground. The limited data on the lead content of precipitation indicate a high variability.

Wet deposition in rural and remote areas can often indicate regional or global processes that remove lead from the atmosphere. Talbot and Andren (1983) measured lead in air and rain at a semiremote site in Wisconsin. They found that wet deposition appeared to represent 80 percent of the total deposition of lead, and the total atmospheric flux of lead was 8 mg/m^2/yr. There was a sharp increase in lead deposition during the summer months.

Deposition to a snowpack can be informative if the measurement correctly samples lead that was deposited with the snow during a period of no snowmelt (Barrie and Vet, 1984). These authors reported deposition of 1.8 mg/m^2/yr to a snowpack of the East Canadian shield. Although they meticulously avoided collecting melted snow, it was not clear how they accounted for dry deposition deposited between periods of snowfall.

A study of cloud droplet capture by vegetation (Lovett, 1984) suggests a possible mechanism of deposition not included in wet or dry deposition. Although data on lead are not available, the mass transfer of water by this mechanism (0.01 cm^3/hr) suggests that, at a concentration of 1 µg Pb/kg, the flux of lead could be 0.01 ng/cm^2 for each hour of cloud droplet exposure.

Lazrus et al. (1970) sampled precipitation at 32 U.S. stations and found a correlation between gasoline used and lead concentrations in rainfall in each area. Similarly, there is probably an inverse correlation between lead concentration in rainfall and distance from large stationary point sources. The authors pointed out that at least twice as much lead is found in precipitation as in water supplies, implying the existence of a process by which lead is removed from water in the soil after precipitation reaches the ground. Russian studies (Konovalov et al., 1966) point to the insolubility of lead compounds in surface waters and suggest removal by natural sedimentation and filtration.

Lindberg et al. (1979) evaluated the deposition of Pb by wet and dry processes in a study at Walker Branch Watershed in eastern Tennessee during the period 1976-1977. The mean annual precipitation in the area is approximately 140 cm. Results for a typical year are reported in Table 6-5. Wet deposition was estimated to contribute approximately 50 percent of the total atmospheric input during this period, but on a seasonal basis ranged from 20 percent to 60 percent of total deposition. Further details on these studies have been published (Lindberg et al., 1982; Lindberg, 1982).

6.4.3 Global Budget of Atmospheric Lead

The geochemical mass balance of lead in the atmosphere may be determined on a global basis from quantitative estimates of inputs and outputs. Inputs are from natural and anthropogenic emissions described in Section 5.2 and 5.3. They amount to 450,000-475,000 metric tons annually (Nriagu, 1979). This simple procedure is an informative exercise that shows whether the observed emission rates and deposition rates can, by making a minimum number of reasonable assumptions, be brought into arithmetic balance. Each assumption can be tested independently, within the constraints of the overall model. For example, Table 6-6 assumes an average concentration of 0.4 µg Pb/kg precipitation. The total mass of rain and snowfall is 5.2×10^{17} kg/yr, so the amount of lead removed by wet deposition is approximately 208,000 t/yr. The average concentration of lead in precipitation cannot be greater than 0.8 µg/kg (although values higher than this are commonly found in the scientific literature), since this would exceed the estimates of global emissions. Furthermore, a value this high would preclude dry deposition. For dry deposition, a crude estimate may be derived by dividing the surface of the earth into three major vegetation types based on surface roughness or LAI. Oceans, polar regions, and deserts have a very low surface roughness and can be assigned a deposition

TABLE 6-5. ANNUAL AND SEASONAL DEPOSITION OF Pb AT WALKER BRANCH WATERSHED,[a] mg/m^2

Period[b]	Atmospheric deposition of Pb	
	Wet	Dry
MEAN Daily deposition	1.9 x 01-2	2.2 x 10-2
Winter	2.4	1.8
Spring	0.3	1.5
Summer	2.7	3.1
Fall	1.6	1.6
Total year	7.0	8.0
Mean daily deposition	1.9×10^{-2}	2.2×10^{-2}

[a] Calculated for a typical year from data collected during 1976-1977.

[b] Winter = November-February, Spring = March and April, Summer = May-August, Fall = September and October.

Source: Lindberg et al., 1979.

TABLE 6-6. ESTIMATED GLOBAL DEPOSITION OF ATMOSPHERIC LEAD

	Mass of water, 10^{17} kg/yr	Lead concentration, 10^{-6} g/kg	Lead deposition, 10^6 kg/yr
Wet			
To oceans	4.1	0.4	164
To continents	1.1	0.4	44
		Total wet:	208
Dry	Area, 10^{12} m^2	Deposition rate, 10^{-3} g/m^2·yr	Deposition, 10^6 kg/yr
To oceans, ice caps, deserts	405	0.22	89
Grassland, agricultural areas, and tundra	46	0.71	33
Forests	59	1.5	80
		Total dry:	202
		Total wet:	208
		Global:	410

Source: This report.

velocity of 0.01 cm/sec, which gives a flux of 0.2 $\mu g/m^2 \cdot yr$, assuming 75 ng Pb/m^3 air concentration. Grasslands, tundra, and other areas of low-lying vegetation have a somewhat higher deposition velocity; forests would have the highest. Values of 0.3 and 0.65 can be assigned to these two vegetation types, based on the data of Davidson et al. (1982). Whittaker (1975) lists the global surface area of each of the three types as 405, 46, and 59 x 10^{12} m^2, respectively. In the absence of data on the global distribution of air concentrations of lead, an average of 0.075 $\mu g/m^3$ is assumed. Multiplying air concentration by deposition velocity gives the deposition flux for each surface roughness type shown on Table 6-6. The combined wet and dry deposition is 410,000 metric tons, which compares favorably with the estimated 450,000-475,000 metric tons of emissions.

The data used above are not held to be absolutely firm. Certainly, more refined estimates of air concentrations and deposition velocities can be made in the future. On the other hand, the calculations above show some published calculations to be unreasonable. In particular, if the values for lead in rain (36 $\mu g/kg$) reported by Lazrus et al. (1970) were applied to this global model, more than 50 times the total global emissions would be required for mass balance. Likewise, deposition fluxes of 0.95 $\mu g/cm^2 \cdot yr$ reported by Jaworowski et al. (1981) would account for 10 times global emissions. Chemical mass balance budgets are an effective aid to evaluating reports of environmental lead data.

6.5 TRANSFORMATION AND TRANSPORT IN OTHER ENVIRONMENTAL MEDIA

6.5.1 Soil

The accumulation of lead in soils is primarily a function of the rate of deposition, since most lead is retained by the soil and very little passes through into surface or ground water. The wet and dry deposition rates discussed in Section 6.4 would apply provided the surface roughness and location (urban, rural, remote) are considered. A value of 8 mg/m^2 yr such as that measured by Talbot and Andren (1983) in a semiremote location in Wisconsin converts to 0.8 $\mu g/cm^2$ yr. It is difficult to generalize on the depth of penetration of lead in undisturbed soils, but if it is assumed that most of the lead is retained in the upper 5 cm (Reaves and Berrow, 1984; Garcia-Miragaya, 1984), then the accumulation rate of 0.8 $\mu g/cm^2$ yr becomes 0.16 $\mu g/cm^3$ yr, or 0.16 $\mu g/g$ if a density of 1 is assumed for soil. Ewing and Pearson (1974) reported an accumulation of 13 $\mu g/g$ soil from the 1920's to the late 1960's, or an annual rate of about 0.26 $\mu g/g$ in a rural setting. It should be noted that the atmospheric concentration of lead increased 20-fold during this period (Shirahata et al., 1980). Page and Ganje (1970) found an accumulation of 0.83 $\mu g/g$ during the same time for a site near high traffic density.

These accumulation rates are discussed further in Section 7.2.2.1. Understanding the distinction between atmospheric and natural lead in soil can provide some insight into the mechanisms regulating transport in soil. Of particular importance are solubility and the stability of lead complexes with humic substances.

Soils have both a liquid and solid phase, and trace metals are normally distributed between these two phases. In the liquid phase, metals may exist as free ions or as soluble complexes with organic or inorganic ligands. Organic ligands are typically humic substances such as fulvic or humic acid; inorganic ligands may be iron or manganese hydrous oxides. Since lead rarely occurs as a free ion in the liquid phase (Camerlynck and Kiekens, 1982), its mobility in the soil solution depends on the availability of organic or inorganic ligands. The liquid phase of soil often exists as a thin film of moisture in intimate contact with the solid phase. The availability of metals to plants depends on the equilibrium between the liquid and solid phase.

In the solid phase, metals may be incorporated into crystalline minerals of parent rock material, into secondary clay minerals, or precipitated as insoluble organic or inorganic complexes. They may also be adsorbed onto the surfaces of any of these solid forms. Of these categories, the most mobile form is in the film of moisture surrounding soil particles, where lead can move freely into plant roots or soil microorganisms with dissolved nutrients. The least mobile is parent rock material, where lead may be bound within crystalline structures over geologic periods of time. Intermediate are the lead complexes and precipitates. Transformation from one form to another depends on the chemical environment of the soil. For example, at pH 6-8, insoluble organic-Pb complexes are favored if sufficient organic matter is available; otherwise hydrous oxide complexes may form or the lead may precipitate with the carbonate or phosphate ion. In the pH range of 4-6, the organic-Pb complexes become soluble. Soils outside the pH range of 4-8 are rare. The interconversion between soluble and insoluble organic complexes affects the equilibrium of lead between the liquid and solid phase of soil.

Dong et al. (1985) found that only 0.2 percent of the total lead in soil can be released to solution by physical shaking. Even if 99.99 percent of the total lead in soil is immobilized, 0.01 percent of the total lead in soil can have a significant effect on plants and microorganisms if the soils are heavily contaminated with lead (see Section 8.3.1).

The water soluble and exchangeable (as determined by chemical extraction) forms of metals are the forms generally considered potentially available for plant uptake. It is important not to confuse the term "extractable" with "plant uptake." Lead that can be extracted from soil by chemical treatment may not be taken up by plants, even though the same chemical treatment is known to release other metals to plants. Because little is known of this relationship, lead that is extractable by chemical means normal for other metals is considered only potentially available for plant uptake. Camerlynck and Kiekens (1982) demonstrated that in normal soils, only a small fraction of the total lead is in exchangeable form (about 1 μg/g) and none exists as free lead ions. Of the exchangeable lead, 30 percent existed as stable complexes, 70 percent as labile complexes. The organic content of these soils was low (3.2 percent clay, 8.5 percent silt, 88.3 percent sand). In heavily contaminated soils near a mid-western industrial site, Miller and McFee (1983) found that 77 percent of the lead was in either the exchangeable or organic form, although still none could be found in aqueous solution. Soils had a total lead content from 64 to 360 μg/g and an organic content of 7-16 percent.

There is evidence that atmospheric lead enters the soil system as $PbSO_4$ or is rapidly converted to $PbSO_4$ at the soil surface (Olson and Skogerboe, 1975). Lead sulfate is more soluble than $PbCO_3$ or $Pb_3(PO_4)_2$ and thus could remain mobile if not transformed. Lead could be immobilized by precipitation as less soluble compounds [$PbCO_3$, $Pb_3(PO_4)_2$], by ion exchange with hydrous oxides or clays, or by chelation with humic and fulvic acids. Santillan-Medrano and Jurinak (1975) discussed the possibility that the mobility of lead is regulated by the formation of $Pb(OH)_2$, $Pb_3(PO_4)_2$, $Pb_5(PO_4)_3OH$, and $PbCO_3$. This model, however, did not consider the possible influence of organic matter on lead immobilization. Zimdahl and Skogerboe (1977), on the other hand, found lead varied linearly with cation exchange capacity (CEC) of soil at a given pH, and linearly with pH at a given CEC (Figure 6-9). The relationship between CEC and organic carbon is discussed below.

If surface adsorption on clays plays a major role in lead immobilization, then the capacity to immobilize should vary directly with the surface-to-volume ratio of clay. In two separate experiments using the nitrogen BET method for determining surface area and size fractionation techniques to obtain samples with different surface-to-volume ratios, Zimdahl and Skogerboe (1977) demonstrated that this was not the case. They also showed that precipitation as lead phosphate or lead sulfate is not significant, although carbonate precipitation can be important in soils that are carbonaceous in nature or to which lime ($CaCO_3$) has been added.

Of the two remaining processes, lead immobilization is more strongly correlated with organic chelation than with iron and manganese oxide formation (Zimdahl and Skogerboe, 1977). It is possible, however, that chelation with fulvic and humic acids is catalyzed by the presence of iron and manganese oxides (Saar and Weber, 1982). This would explain the positive correlation for both mechanisms observed by Zimdahl and Skogerboe (1977). The study of Miller and McFee (1983) discussed above indicates that atmospheric lead added to soil is distributed to organic matter (43 percent) and ferro-manganese hydrous oxides (39 percent), with 8 percent found in the exchangeable fraction (determined by chemical extraction) and 10 percent as insoluble precipitates.

If organic chelation is the correct model of lead immobilization in soil, then several features of this model merit further discussion. First, the total capacity of soil to immobilize lead can be predicted from the linear relationship developed by Zimdahl and Skogerboe (1977) (Figure 6-9) based on the equation:

$$N = 2.8 \times 10^{-6} (A) + 1.1 \times 10^{-5} (B) - 4.9 \times 10^{-5} \qquad (6\text{-}2)$$

where N is the saturation capacity of the soil expressed in moles/g soil, A is the CEC of the soil in meq/100 g soil, and B is the pH. Because the CEC of soil is more difficult to determine than total organic carbon, it is useful to define the relationship between CEC and organic content. Pratt (1957) and Klemmedson and Jenny (1966) found a linear correlation between CEC and organic carbon for soils of similar sand, silt, and clay content. The data of Zimdahl and Skogerboe (1977) also show this relationship when grouped by soil type. They show that sandy clay loam with an organic content of 1.5 percent might be expected to have a CEC of 12 meq/100 g. From the equation, the saturation capacity for lead in soil of pH 5.5 would be 45 μmoles/g soil or 9,300 μg/g. The same soil at pH 4.0 would have a total capacity of 5,900 μg/g.

When the soil contains sufficient organic matter, the adsorption of lead and other metals is a function of pH. In reality, it is not the total organic matter but the number of reactive sites that determines metal absorption (Harter, 1983), and there appears to be a weak relationship between cation exchange capacity and metal retention. In competition with other metals, lead would normally be more strongly favored for retention, in accordance with the Irving-Williams series (Irving and Williams, 1953). Gamble et al. (1983) have shown that the Irving-Williams series becomes somewhat distorted when the binding sites are chemically dissimilar.

The soil humus model also facilitates the calculation of lead in soil moisture using values available in the literature for conditional stability constants with fulvic acid (FA). The term conditional is used to specify that the stability constants are specific for the conditions of the reaction. Conditional stability constants for humic acid (HA) and FA are comparable. The values reported for log K are linear in the pH range of 3 to 6 (Buffle and Greter, 1979; Buffle et al., 1976; Greter et al., 1979), so that interpolations in the critical range of pH 4 to 5.5 are possible. Thus, at pH 4.5, the ratio of complexed lead to ionic lead is expected to be 3.8×10^3. For soils of 100 μg/g, the ionic lead in soil moisture solution would be 0.03 μg/g. The significance of this ratio is discussed in Section 8.3.1.1.

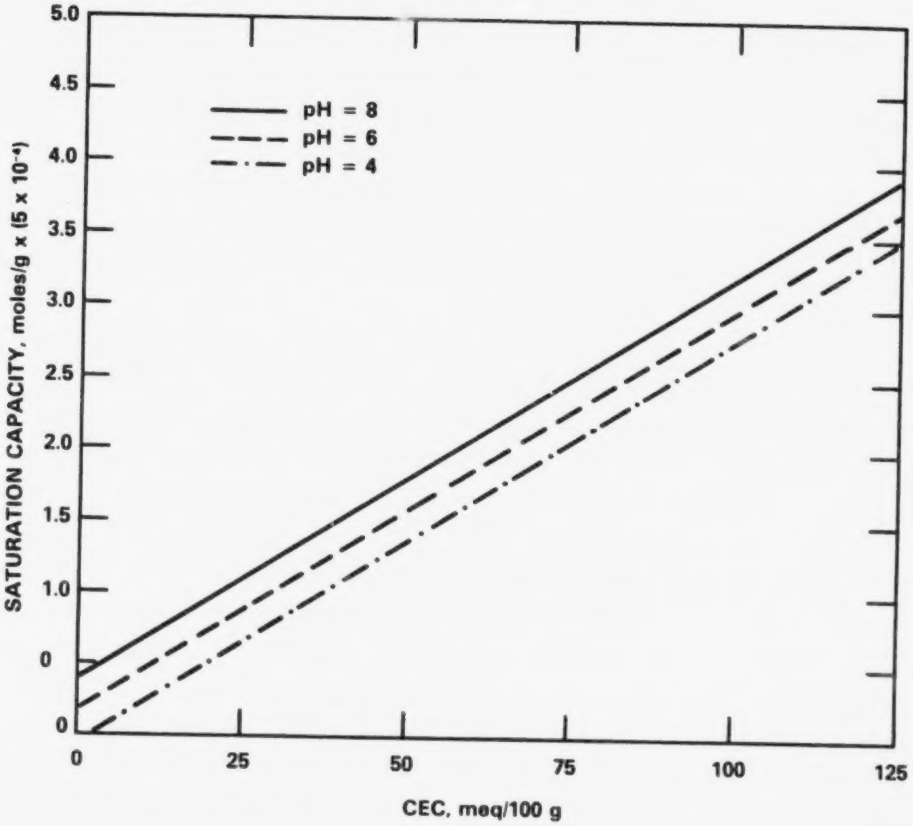

Figure 6-9. Variation of lead saturation capacity with cation exchange capacity (CEC) in soil at selected pH values.

Source: Data from Zimdahl and Skogerboe (1977).

It is also important to consider the stability constant of the Pb-FA complex relative to other metals. Schnitzer and Hansen (1970) showed that at pH 3, Fe^{3+} is the most stable in the sequence $Fe^{3+} > Al^{3+} > Cu^{2+} > Ni^{2+} > Co^{2+} > Pb^{2+} > Ca^{2+} > Zn^{2+} > Mn^{2+} > Mg^{2+}$. At pH 5, this sequence becomes $Ni^{2+} = Co^{2+} > Pb^{2+} > Cu^{2+} > Zn^{2+} = Mn^{2+} > Ca^{2+} > Mg^{2+}$. This means that at normal soil pH levels of 4.5-8, lead is bound to FA and HA in preference to many other metals that are known plant nutrients (Zn, Mn, Ca, and Mg). Furthermore, if lead displaces iron in this scheme, an important function of FA may be inhibited at near saturation capacity (above 6000 µg Pb/g.) Fulvic acid is believed to play a role in the weathering of parent rock material by the removal of iron from the crystalline structure of the minerals, causing the rock to weather more rapidly. In the absence of this process, the weathering of parent rock material and the subsequent release of nutrients to soil would proceed more slowly. Bizri et al. (1984) found stability constants for humic substances were $\log K_1 = 4.2$ and $\log K_2 = 3.7$. For humic materials in aquatic systems, Alberts and Giesy (1983) reported conditional stability constants of $\log K_1 = 5.09$ and $\log K_2 = 2.00$.

6.5.2 Water

6.5.2.1 Inorganic. The chemistry of lead in an aqueous solution is highly complex because the element can be found in a multiplicity of forms. Hem and Durum (1973) have reviewed the chemistry of lead in water in detail; the aspects of aqueous lead chemistry that are germane to this document are discussed in Section 3.3.

Lead in ore deposits does not pass easily to ground or surface water. Any lead dissolved from primary lead sulfide ore tends to combine with carbonate or sulfate ions to (1) form insoluble lead carbonate or lead sulfate, or (2) be absorbed by ferric hydroxide (Lovering, 1976). An outstanding characteristic of lead is its tendency to form compounds of low solubility with the major anions of natural water. Hydroxide, carbonate, sulfide, and more rarely sulfate may act as solubility controls in precipitating lead from water. The amount of lead that can remain in solution is a function of the pH of the water and the dissolved salt content. Equilibrium calculations show that at pH > 5.4, the total solubility of lead is about 30 µg/l in hard water and about 500 µg/l in soft water (Davies and Everhart, 1973). Lead sulfate is present in soft water and limits the lead concentration in solution. Above pH 5.4, $PbCO_3$ and $Pb_2(OH)_2CO_3$ limit the concentration. The carbonate concentration is in turn dependent on the partial pressure of CO_2 as well as the pH. Calculations by Hem and Durum (1973) show that many river waters in the United States have lead concentrations near the solubility limits imposed by their pH levels and contents of dissolved CO_2. Because of the influence of temperature on the solubility of CO_2, observed lead concentrations may vary significantly from theoretically calculated ones.

Concentrations as high as 330 μg/l could be stable in water with pH near 6.5 and an alkalinity of about 25 mg bicarbonate ion/l of water. Water having these properties is common in runoff areas of New York State and New England; hence, the potential for high lead concentrations exists there. In other areas, the average pH and alkalinity are so high that maximum concentrations of lead of about 1 μg/l could be retained in solutions at equilibrium (Lovering, 1976).

A significant fraction of the lead carried by river water may be in an undissolved state. This insoluble lead can consist of colloidal particles or larger undissolved particles of lead carbonate, -oxide, -hydroxide, or other lead compounds incorporated in other components of particulate lead from runoff; it may occur either as sorbed ions or surface coatings on sediment mineral particles or be carried as a part of suspended living or nonliving organic matter (Lovering, 1976). A laboratory study by Hem (1976) of sorption of lead by cation exchange indicated that a major part of the lead in stream water may be adsorbed on suspended sediment. Figure 6-10 illustrates the distribution of lead outputs between filtrate and solids in water from both urban and rural streams, as reported by Getz et al. (1977). The majority of lead output is associated with suspended solids in both urban and rural streams, with very little dissolved in the filtrate. The ratio of lead in suspended solids to lead in filtrate varies from 4:1 in rural streams to 27:1 in urban streams.

Soluble lead is operationally defined as that fraction which is separated from the insoluble fraction by filtration. However, most filtration techniques do not remove all colloidal particles. Upon acidification of the filtered sample, which is usually done to preserve it before analysis, the colloidal material that passed through the filter is dissolved and is reported as dissolved lead. Because the lead in rainfall can be mainly particulate, it is necessary to obtain more information on the amounts of insoluble lead (Lovering, 1976) before a valid estimate can be obtained of the effectiveness of runoff in transporting lead away from areas where it has been deposited by wet and dry deposition.

6.5.2.2 Organic. The bulk of organic compounds in surface waters originates from natural sources (Neubecker and Allen, 1983). The humic and fulvic acids that are primary complexing agents in soils are also found in surface waters at concentrations from 1-5 mg/l, occasionally exceeding 10 mg/l (Steelink, 1977), and have approximately the same chemical characteristics (Reuter and Perdue, 1977). The most common anthropogenic organic compounds are nitrilotriacetonitrile (NTA) and ethylenediaminetetraacetic acid (EDTA) (Neubecker and Allen, 1983). There are many other organic compounds such as oils, plasticizers, and polymers discharged from manufacturing processes that may complex with lead.

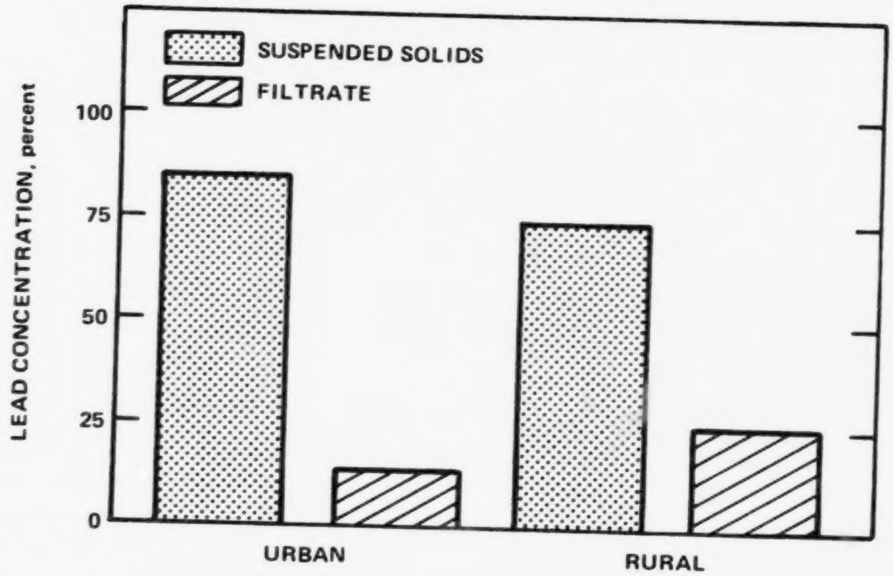

Figure 6-10. Lead distribution between filtrate and suspended solids in stream water from urban and rural compartments.

Source: Getz et al. (1977).

The presence of fulvic acid in water has been shown to increase the rate of solution of lead sulfide 10 to 60 times over that of a water solution at the same pH that did not contain fulvic acid (Bondarenko, 1968; Lovering, 1976). At pH values near 7, soluble Pb-FA complexes are present in solution. At initial pH values between 7.4 and about 9, the Pb-FA complexes are partially decomposed, and lead hydroxide and carbonate are precipitated. At initial pH values of about 10, the Pb-FA complexes again increase. This increase is attributed to dissociation of phenolic groups at high pH values, which increases the complexing capacity of the FA. But it also may be due to the formation of soluble lead-hydroxyl complexes.

Bëijer and Jernelov (1984) review the evidence for the microbial methylation of lead in aquatic systems. The transformation of inorganic lead, especially in sediment, to tetramethyllead (TML) has been observed and biomethylation has been postulated (Schmidt and Huber, 1976; Wong et al., 1975). Reisinger et al. (1981) have reported extensive studies of the methylation of lead in the presence of numerous bacterial species known to alkylate mercury and other heavy metals. In these experiments no biological methylation of lead was found under any condition. Chemical alkylation from methylcobalamine was found to occur in the presence of sulfide or of aluminum ion; chemical methylation was independent of the presence of bacteria.

Jarvie et al. (1975, 1981) have recently shown that tetraalkyllead compounds are unstable in water. Small amounts of Ca^{2+} and Fe^{2+} ions and sunlight have been shown to cause decomposition of TEL over time periods of 5-50 days. The only product detected was triethyllead, which appears to be considerably more stable than the TEL. Tetramethyllead is decomposed much more rapidly than TEL in water, to form the trimethyl lead ion. Initial concentrations of 10^{-4} molar were reduced by one order of magnitude either in the dark or light in one day. Tetramethyllead was virtually undetectable after 21 days. Apparently, chemical methylation of lead to the trialkyllead cation does occur in some water systems, but evolution of TML appears insignificant.

Lead occurs in riverine and estuarial waters and alluvial deposits. Laxen and Harrison (1977) and Harrison and Laxen (1981) found large concentrations of lead (~1 mg/l) in rainwater runoff from a roadway; but only 5-10 percent of this is soluble in water. Concentrations of lead in ground water appear to decrease logarithmically with distance from a roadway. Rainwater runoff has been found to be an important transport mechanism in the removal of lead from a roadway surface in a number of studies (Bryan, 1974; Harrison and Laxon, 1981; Hedley and Lockley, 1975; Laxen and Harrison, 1977). Apparently, only a light rainfall, 2-3 mm, is sufficient to remove 90 percent of the lead from the road surface to surrounding soil and to waterways (Laxen and Harrison, 1977).

The Applied Geochemistry Research Group (1978) has reported elevated lead concentrations (40 μg/g and above) in about 30 percent of stream bed sediment samples from England and Wales in a study of 50,000 such samples. Abdullah and Royle (1973) have reported lead levels in coastal areas of the Irish sea of 400 μg/g and higher.

Evidence for the sedimentation of lead in freshwater streams may be found in several reports. Laxen and Harrison (1983) found that lead in the effluent of a lead-acid battery plant near Manchester, England, changed drastically in particle size. In the plant effluent, 53 percent of the lead was on particles smaller than 0.015 μm and 43 percent on particles greater than 1 μm. Just downstream of the plant, 91 percent of the lead was on particles greater than 1 μm and only 1 percent on particles smaller than 0.015 μm. Under these conditions, lead formed or attached to large particles at a rate exceeding that of Cd, Cu, Fe or Mn.

The lead concentrations in off-shore sediments often show a marked increase corresponding to anthropogenic activity in the region (Section 5.1). Rippey et al. (1982) found such increases recorded in the sediments of Lough Neagh, Northern Ireland, beginning during the 1600's and increasing during the late 1800's. Corresponding increases were also observed for Cr, Cu, Zn, Hg, P, and Ni. For lead, the authors found an average anthropogenic flux of 72 $mg/m^2 \cdot yr$, of which 27 $mg/m^2 \cdot yr$ could be attributed to direct atmospheric deposition. Prior to 1650, the total flux was 12 $mg/m^2 \cdot yr$, so there has been a 6-fold increase since that time.

Ng and Patterson (1982) found prehistoric fluxes of 1-7 mg $Pb/m^2 \cdot yr$ in three offshore basins in southern California, which have now increased 3 to 9-fold to 11-21 $mg/m^2 \cdot yr$. Much of this lead is deposited directly from sewage outfalls, although at least 25 percent probably comes from the atmosphere.

6.5.3 Vegetation Surfaces

The deposition of lead on the leaf surfaces of plants where the particles are often retained for a long time must also be considered (Dedolph et al., 1970; Page et al., 1971; Schuck and Locke, 1970). Many studies have shown that plants near roadways exhibit considerably higher levels of lead than those further away. In most instances, the higher concentrations were due to lead particle deposition on plant surfaces (Schuck and Locke, 1970). Studies have shown that particles deposited on plant surfaces are difficult to remove by typical kitchen washing techniques. (Arvik and Zimdahl, 1974; Page et al., 1971; Lagerwerff et al., 1973). Leaves with pubescent surfaces seem able to retain particles via an electrostatic mechanism. Other types of leaves are covered with a cuticular wax physically suitable for retaining particles. Rainfall does not remove all of the particles on the leaf surface. It appears that there is a buildup with time of surface deposition on leafy vegetation. Animals consuming the leafy portions of such plants can certainly be exposed to higher than normal levels of lead.

The uptake of soluble lead by aquatic plants can be an important mechanism for depleting lead concentrations in downstream waterways. Gale and Wixson (1979) have studied the influence of algae, cattails, and other aquatic plants on lead and zinc levels in wastewater in the New Lead Belt of Missouri. These authors report that mineral particles become trapped by roots, stems, and filaments of aquatic plants. Numerous anionic sites on and within cell walls participate in cation exchange, replacing metals such as lead with Na^+, K^+, and H^+ ions. Mineralization of lead in these Missouri waters may also be promoted by water alkalinity. However, construction of stream meanders and settling ponds have greatly reduced downstream water concentrations of lead, mainly because of absorption in aquatic plants (Gale and Wixson, 1979).

6.6 SUMMARY

From the source of emission to the site of deposition, lead particles are dispersed by the flow of the airstream, transformed by physical and chemical processes, and removed from the atmosphere by wet or dry deposition. Under the simplest of conditions (smooth, flat terrain), the dispersion of lead particles has been modeled and can be predicted (Benarie, 1980). Dispersion modeling in complex terrains is still under development and these models have not been evaluated (Kotake and Sano, 1981).

Air lead concentrations decrease logarithmically away from roadways (Edwards, 1976) and smelters (Roberts et al., 1974). Within urban regions, air concentrations decrease from the central business district to the outlying residential areas by a factor of 2-3. From urban to rural areas, air concentrations decrease from 1-2 $\mu g/m^3$ down to 0.1-0.5 $\mu g/m^3$ (Chapter 7). This decrease is caused by dilution with clean air and removal by deposition. During dispersion to remote areas, concentrations decrease to 0.01 $\mu g/m^3$ in the United States (Elias and Davidson, 1980), to 0.001 $\mu g/m^3$ in the Atlantic Ocean (Duce et al., 1975), and to 0.000076 $\mu g/m^3$ in Antarctica (Maenhaut et al., 1979).

Physical transformations of lead particles cause a shift in the particle size distribution. The bimodal distribution of large and small particles normally found near the roadway changes with time and distance to a single mode of intermediate sized particles (Huntzicker et al., 1975). This is probably because large particles deposit near roadways and small particles agglomerate to medium sized particles with an MMAD of about 0.2-0.3 μm.

Particles transform chemically from lead halides to lead sulfates and oxides. Organolead compounds constitute 1-6 percent of the total airborne lead in ambient urban air (Harrison et al., 1979).

On a regional or global basis, wet deposition accounts for about half of the removal of lead particles from the atmosphere. The other half of the atmospheric lead is removed by dry deposition. Mechanisms may be gravitational for large particles or a combination of gravitational and wind-related mechanisms for small particles (Elias and Davidson, 1980). Models of dry deposition predict deposition velocities as a function of particle size, windspeed, and surface roughness. Because of their large surface area/ground area ratio, grasslands, croplands, and forested areas receive the bulk of dry deposited particles over continental areas.

Lead enters soil as a moderately insoluble lead sulfate and is immobilized by complexation with humic and fulvic acids. This immobilization is a function of pH and the concentration of humic substances. At low pH (~4) and low organic content (<5 percent), immobilization of lead in soil may be limited to a few hundred µg/g (Zimdahl and Skogerboe, 1977), but at 20 percent organic content and pH 6, 10,000 µg Pb/g soil may be found.

In natural waters, lead may precipitate as lead sulfate or carbonate, or it may form a complex with ferric hydroxide (Lovering, 1976). The solubility of lead in water is a function of pH and hardness (a combination of Ca and Mg content). Below pH 5.4, concentrations of dissolved lead may vary from 30 µg/l in hard water to 500 µg/l in soft water at saturation (Lovering, 1976).

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7. ENVIRONMENTAL CONCENTRATIONS AND POTENTIAL PATHWAYS TO HUMAN EXPOSURE

7.1 INTRODUCTION

In general, typical levels of human lead exposure may be attributed to four components of the human environment: food, inhaled air, dusts of various types, and drinking water. This chapter presents information on the ranges and temporal trends of concentrations in ambient air, soil, and natural waters, and discusses the pathways from each source to food, inhaled air, dust, and drinking water. The ultimate goal is to quantify the contribution of anthropogenic lead to each source and the contribution of each source to the total lead consumed by humans. These sources and pathways of human lead exposure are diagrammed in Figure 7-1.

Chapters 5 and 6 discuss the emission, transport, and deposition of lead in ambient air. Some information is also presented in Chapter 6 on the accumulation of lead in soil and on plant surfaces. Because this accumulation is at the beginning of the human food chain, it is critical to understand the relationship between this lead and lead in the human diet. It is also important where possible to project temporal trends.

In this chapter, a baseline level of potential human exposure is determined equivalent to that for a normal adult eating a typical diet and living in a non-urban community. This baseline exposure is deemed to be unavoidable by any reasonable means. Beyond this level, additive exposure factors can be determined for other environments (e.g., urban, occupational, smelter communities), for certain habits and activities (e.g., pica, smoking, drinking, and hobbies), and for variations due to age, sex, or socioeconomic status.

7.2 ENVIRONMENTAL CONCENTRATIONS

Quantifying human exposure to lead requires an understanding of ambient lead levels in environmental media. Of particular importance are lead concentrations in ambient air, soil, and surface or ground water. The following sections discuss environmental lead concentrations in each of these media in the context of anthropogenic vs. natural origin, and the contribution of each to potential human exposure.

7.2.1 Ambient Air

Ambient airborne lead concentrations may influence human exposure through direct inhalation of lead-containing particles and through ingestion of lead that has been deposited from the air onto surfaces. Although a plethora of data on airborne lead is now available, our

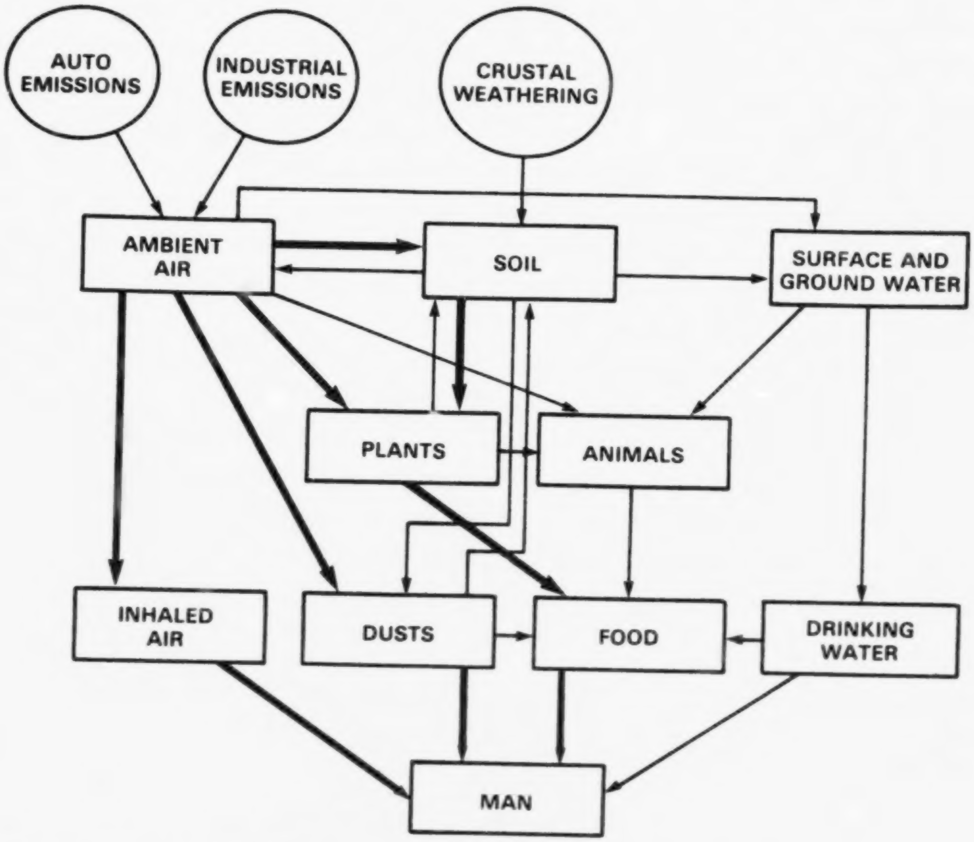
AUTO EMISSIONS
INDUSTRIAL EMISSIONS
CRUSTAL WEATHERING
AMBIENT AIR
SOIL
SURFACE AND GROUND WATER
PLANTS
ANIMALS
INHALED AIR
DUSTS
FOOD
DRINKING WATER
MAN

Figure 7-1. Principal pathways of lead from the environment to human consumption. Heavy arrows are those pathways discussed in greatest detail in this chapter.

understanding of the pathways to human exposure is far from complete because most ambient measurements were not taken in conjunction with studies of the concentrations of lead in man or in components of his food chain. However, that is the context in which these studies must now be interpreted to shed the most light possible on the concentrations likely to be encountered in various environmental settings.

The most complete set of data on ambient air concentrations may be extracted from the National Filter Analysis Network (NFAN) and its predecessors (see Section 4.2.1). These data, which are primarily for urban regions, have been supplemented with published data from rural and remote regions of the United States. Because some stations in the network have been in place for about 15 years, information on temporal trends is available but sporadic. Ambient air concentrations in the United States are comparable to other industrialized nations. In remote regions of the world, air concentrations are two or three orders of magnitude lower, lending credence to estimates of the concentration of natural lead in the atmosphere. In the context of the NFAN data base, the conditions are considered that modify ambient air, as measured by the monitoring networks, to air as inhaled by humans. Specifically, these conditions are changes in particle size distributions, changes with vertical distance above ground, and differences between indoor and outdoor concentrations.

7.2.1.1 Total Airborne Lead Concentrations. A thorough understanding of human exposure to airborne lead requires detailed knowledge of spatial and temporal variations in ambient concentrations. The wide range of concentrations is apparent from Table 7-1, which summarizes data obtained from numerous independent measurements, and Tables 7-2 and 7-3, which show air concentrations in specific locations throughout the United States. Concentrations vary from 0.000076 μg/m^3 in remote areas to over 13 μg/m^3 near sources such as smelters. Many of the remote areas are far from human habitation and therefore do not reflect human exposure. However, a few of the regions characterized by low lead concentrations are populated by individuals with primitive lifestyles; these data provide baseline airborne lead data to which modern American lead exposures can be compared. Examples include some of the data from South America and the data from Nepal. A more extensive review of atmospheric lead in remote areas has been compiled by Wiersma and Davidson (1984).

Urban, rural, and remote airborne lead concentrations in Table 7-1 suggest that human exposure to lead has increased as the use of lead in inhabited areas has increased. This is consistent with published results of retrospective human exposure studies. For example, Ericson et al. (1979) have analyzed the teeth and bones of Peruvians buried 1600 years ago. Based on their data, they estimate that the skeletons of present-day American and British adults contain roughly 500 times the amount of lead that would occur naturally in the absence of widespread anthropogenic lead emissions. Grandjean et al. (1979) and Shapiro et al. (1980)

TABLE 7-1. ATMOSPHERIC LEAD IN URBAN, RURAL, AND REMOTE AREAS OF THE WORLD

Location	Sampling period	Lead conc., (μg/m^3)	Reference
Urban			
New York	1978-79	1.1	NEDS, 1982
Boston	1978-79	0.8	NEDS, 1982
St. Louis	1973	1.1	NEDS, 1982
Houston	1978-79	0.9	NEDS, 1982
Chicago	1979	0.8	NEDS, 1982
Los Angeles	1978-79	1.4	NEDS, 1982
Ottowa	1975	1.3	NAPS, 1971-1976
Toronto	1975	1.3	NAPS, 1971-1976
Montreal	1975	2.0	NAPS, 1971-1976
Brussels	1978	0.5	Roels et al., 1980
Turin	1974-79	4.5	Facchetti and Geiss, 1982
Riyadh, Saudi Arabia	1983	5.5	El-Shobokshy, 1984
Rural			
New York Bight	1974	0.13	Duce et al., 1975
United Kingdom	1972	0.13	Cawse, 1974
Italy	1976-80	0.33	Facchetti and Geiss, 1982
Belgium	1978	0.37	Roels et al., 1980
Illinois	1973-74	0.23	Hudson et al., 1975
Remote			
White Mtn., CA	1969-70	0.008	Chow et al., 1972
High Sierra, CA	1976-77	0.021	Elias and Davidson, 1980
Olympic Nat. Park, WA	1980	0.0022	Davidson et al., 1982
Great Smoky Mtns. Nat. Park, TN	1979	0.015	Davidson et al., 1985
Glacier Nat. Park, MT	1981	0.0046	Davidson et al., 1985
South Pole	1974	0.000076	Maenhaut et al., 1979
Thule, Greenland	1965	0.0005	Murozumi et al., 1969
Thule, Greenland	1978-79	0.008	Heidam, 1983
Prins Christiansund, Greenland	1978-79	0.018	Heidam, 1983
Dye 3, Greenland	1979	0.00015	Davidson et al., 1981c
Eniwetok, Pacific Ocean	1979	0.00017	Settle and Patterson, 1982
Kumjung, Nepal	1979	0.00086	Davidson et al., 1981b
Bermuda	1973-75	0.0041	Duce et al., 1976
Abastumani Mtns. USSR	1979	0.019	Dzubay et al., 1984

report lead levels in teeth and bones of contemporary populations to be elevated 100-fold over levels in ancient Nubians buried before 750 A.D. On the other hand, Barry and Connolly (1981) report excessive lead concentrations in buried medieval English skeletons; one cannot discount the possibility that the lead was absorbed into the skeletons from the surrounding soil.

The remote area concentrations reported in Table 7-1 do not necessarily reflect natural, preindustrial lead. Murozumi et al. (1969) measured a 200-fold increase over the past 3000 years in the lead content of Greenland snow, confirmed by Ng and Patterson (1981). In the opinion of these authors, this lead originates in populated mid-latitude regions, and is transported over thousands of kilometers through the atmosphere to the Arctic. All of the concentrations in Table 7-1, including values for remote areas, have been influenced by anthropogenic lead emissions.

Studies referenced in Table 7-1 are limited in that the procedures for determining the quality of the data are generally not reported. In contrast, the two principal airborne lead data bases described in Section 4.2.1 include measurements subjected to documented quality assurance procedures. The U.S. Environmental Protection Agency's National Filter Analysis Network (NFAN) provides comprehensive nationwide data on long-term trends. The second data base, EPA's National Aerometric Data Bank, contains information contributed by state and local agencies, which monitor compliance with the current ambient airborne standard for lead (1.5 $\mu g/m^3$ averaged over a calendar quarter) promulgated in 1978.

7.2.1.1.1 Distribution of air lead in the United States. Figure 7-2 categorizes the urban sites with valid annual averages (4 valid quarters) into several annual average concentration ranges (Akland, 1976; Shearer et al. 1972; U.S. Environmental Protection Agency, 1978, 1979; Quarterly averages of lead from NFAN, 1982). Nearly all of the sites reported annual averages below 1.0 $\mu g/m^3$. Although the decreasing number of monitoring stations in service in recent years could account for some of the shift in averages toward lower concentrations, trends at individual urban stations, discussed below, confirm the apparent national trend of decreasing lead concentration.

The data from these networks show both the maximum quarterly average to reflect compliance of the station to the ambient airborne standard (1.5 $\mu g/m^3$), and quarterly averages to show trends at a particular location. Valid quarterly averages must include at least five 24-hour sampling periods evenly spaced throughout the quarter. The number of stations complying with the standard has increased, the quarterly averages have decreased, and the maximum 24-hour values appear to be smaller since 1977.

Long-term trends and seasonal variations in airborne lead levels at urban sites can be seen in Figure 7-3. The 10th, 50th, and 90th percentile concentrations are graphed, using quarterly composite and quarterly average data from an original group of 92 urban stations (1965-1974) updated with data for 1975 through 1980. Note that maximum lead concentrations

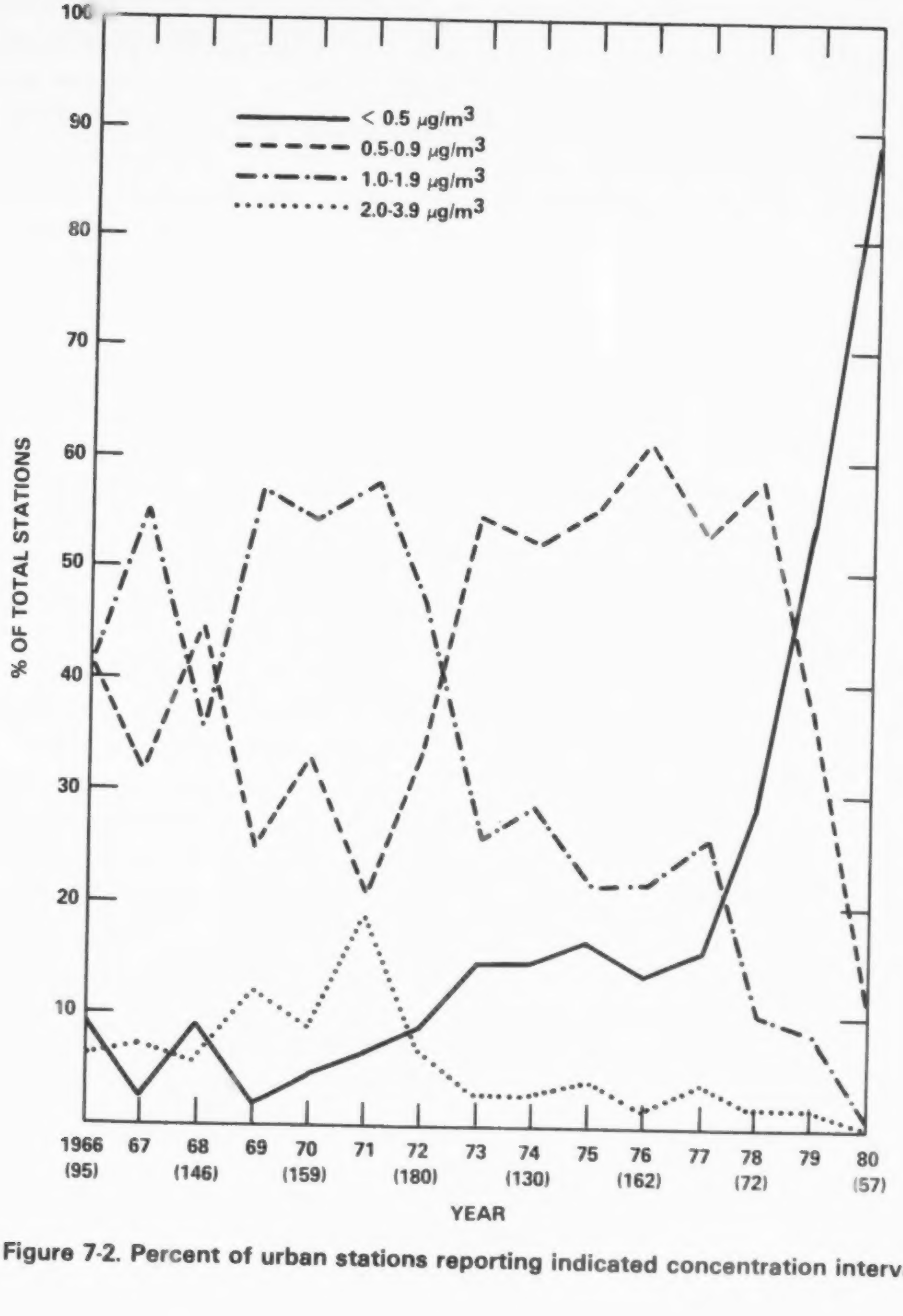

Figure 7-2. Percent of urban stations reporting indicated concentration interval.

typically occur in the winter, while minima occur in the summer. In contrast, automotive emissions of lead would be expected to be greater in the summer for two reasons: (1) gasoline usage is higher in the summer; and (2) lead content is raised in summer gasolines to replace some of the more volatile high-octane components that cannot be used in summertime gasolines. Apparently, the troposphere has a greater capacity to disperse submicron particles in the summer than in the winter.

Figure 7-3 also clearly portrays the significant decrease in airborne lead levels over the past decade. This trend is attributed to the decreasing lead content of regular and premium gasoline, and to the increasing usage of unleaded gasoline. The close parallel between these two parameters is discussed in detail in Chapter 5. (See Figure 5-7 and Table 5-5.)

Table 7-2 shows lead concentrations in the atmospheres of several major metropolitan areas of epidemiological interest. Some of the data presented do not meet the stringent requirements for quarterly averages and occasionally there have been changes in site location or sampling methodology. Nevertheless, the data are the best available for reporting the history of lead contamination in these specific urban atmospheres. Further discussions of these data appear in Chapter 11.

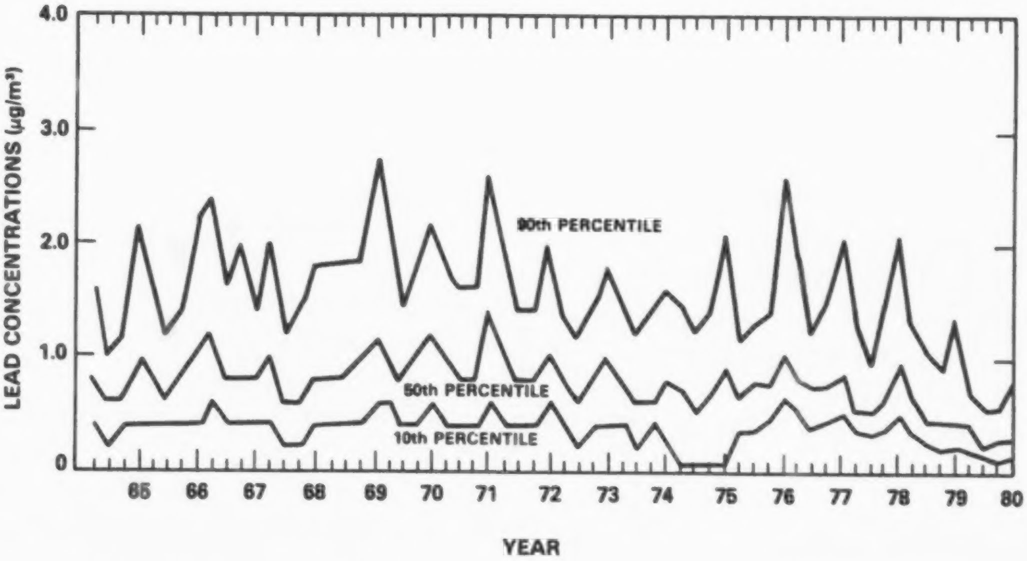

Figure 7-3. Seasonal patterns and trends in quarterly average urban lead concentrations.

7.2.1.1.2 Global distributions of air lead. Other industrialized nations have maintained networks for monitoring atmospheric lead. For example, Kretzschmar et al. (1980) reported trends from 1972 to 1977 in a 15-station network in Belgium. Annual averages ranged from 0.16 μg/m^3 at rural sites to 1.2 μg/m^3 near the center of Antwerp. All urban areas showed a maximum near the center of the city, with lead concentrations decreasing outward. The rural background levels appeared to range from 0.1 to 0.3 μg/m^3. Representative data from other nations appear in Table 7-1.

7.2.1.1.3 Natural concentrations of lead in air. There are no direct measurements of prehistoric natural concentrations of lead in air. Air lead concentrations that existed in prehistoric times must be inferred from available data. Table 7-1 lists several values for remote areas of the world, the lowest of which is 0.000076 μg/m^3 at the South Pole (Maenhaut et al., 1979). Two other reports show comparable values: 0.00017 μg/m^3 at Eniwetok in the Pacific Ocean (Settle and Patterson, 1982) and 0.00015 at Dye 3 in Greenland (Davidson et al., 1981a). Since each of these studies reported some anthropogenic influence, it may be assumed that natural lead concentrations are somewhat lower than these measured values.

Another approach to determining natural concentrations is to estimate global emissions from natural sources. Nriagu (1979) estimated emissions at 24.5 x 10^6 kg/yr, whereas Settle and Patterson (1980) estimated a lower value of 2 x 10^6 kg/yr. An average tropospheric volume, to which surface-generated particles are generally confined, is about 2.55 x 10^{10}m^3. Assuming a residence time of 10 days (see Section 6.3), natural lead emissions during this time would be 6.7 x 10^{14} μg. The air lead concentrations would be 0.000263 μg/m^3 using the values of Nriagu (1979) or 0.0000214 μg/m^3 using the data of Settle and Patterson (1980). It seems likely that the concentration of natural lead in the atmosphere is between 0.00002 and 0.00007 μg/m^3. A value of 0.00005 μg/m^3 will be used for calculations regarding the contribution of natural air lead to total human uptake in Section 7.3.1.

7.2.1.2 Compliance with the 1978 Air Quality Standard. Figure 7-4 shows percentile distributions for the maximum average quarterly lead concentrations by year for a select group of 36 sites for which the data are available during the entire time period, 1975-1984. These data show that not only did the composite average maximum average quarterly values decrease during the period 1975-1984, but the maximum average quarterly lead concentrations for all percentiles showed a comparable pattern of decrease. From Figure 7-4, it may be concluded that most stations reported average quarterly lead concentrations below the NAAQS standard of 1.5 μg/m^3. Those that did not are shown on Table 7-3. Table 7-3 lists stations operated by state and local agencies where one or more quarterly averages exceeded 1.0 μg/m^3 or the current standard of 1.5 μg/m^3 from 1979 to 1984. A portion of each agency's compliance monitoring network consists of monitors sited in areas expected to yield high concentrations associated with identifiable sources. In the case of lead, these locations are most likely to be

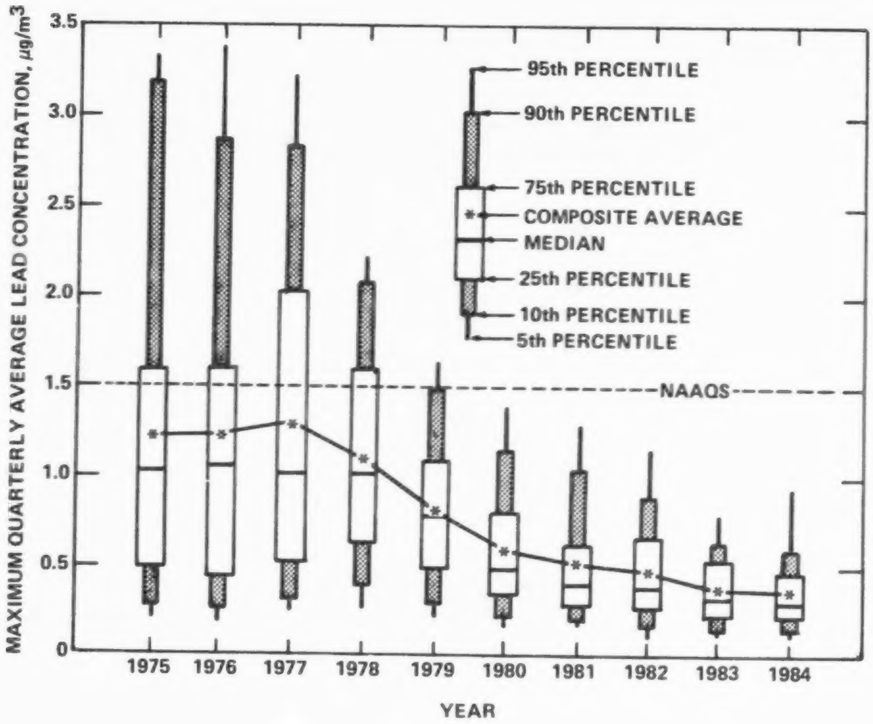

Figure 7-4. Comparison of trends in maximum quarterly average lead concentrations at 36 sites, 1975 - 1984.

Source: U.S. Environmental Protection Agency (1986).

TABLE 7-2. AIR LEAD CONCENTRATIONS IN MAJOR METROPOLITAN AREAS
$\mu g/m^3$ quarterly averages

		Boston MA	New York NY	Phila. PA		Wash. DC	Detroit MI	Chicago IL			Houston TX		Dallas/Ft.Worth TX			Los Angeles CA	
		Station Type															
Year	Quarter	1	1	1	4	1	1	1	2	3	1	4	1	2	4	1	2
1970	1	0.8	1.2			0.9	1.2				1.8		3.8			5.7	3.2
	2		1.5			0.9	1.4				2.0		2.3			3.5	2.2
	3	1.2	1.9				1.4				1.9		2.8			5.1	3.3
	4	1.2	1.4			1.2	1.3				2.5		3.7			3.9	1.9
1971	1		1.6			1.1	1.0				1.9		3.4			6.0	
	2	0.7	1.8			1.3	1.8				1.6		1.8			2.9	
	3					1.3	1.6				1.7		2.5			3.3	
	4		1.7			2.1	2.2				2.7		2.7			6.3	
1972	1	1.0	0.9			1.7					2.3		3.4			3.1	
	2	0.6	1.3			1.2					1.0		1.8			2.0	1.6
	3	2.5	1.0								0.9		2.2			2.6	1.5
	4		1.1			1.1					2.3		2.8			4.7	2.1
1973	1										2.9		1.9			2.7	1.6
	2		0.8								1.8		1.3			2.0	2.5
	3	0.6	1.3								1.7					2.7	
	4		0.9								1.7		1.9				
1974	1		0.5			0.5					1.8		1.3			1.9	1.6
	2	0.9	1.1								2.0	0.6[1]	1.4	0.2[1]		2.0	1.7
	3	1.0	0.9				0.9				1.8	0.6	2.8	0.4		1.4	1.9
	4		0.9				0.9				2.6	0.5	3.3	0.6		3.2	2.6
1975	1	1.2	0.8			1.1	0.8				2.1[1]	0.7	2.9	0.3			1.7
	2	0.6[1]	0.8				0.7				1.7	0.7	2.3	0.3		1.2	1.2
	3	1.0[1]	1.0				1.2				2.1	0.6	3.0	0.4		1.9	1.7
	4	0.9[1]	1.1				1.2				2.4	1.1	2.9	0.5	0.3	3.2	2.2

TABLE 7-2. (continued)

		Boston MA	New York NY	Phila. PA		Wash. DC	Detroit MI	Chicago IL			Houston TX		Dallas/Ft.Worth TX			Los Angeles CA	
		Station Type															
Year	Quarter	1	1	1	4	1	1	1	2	3	1	4	1	2	4	1	2
1976	1										0.8[1]	0.5	0.7[1]	0.3	0.2		
	2					1.2[1]					0.7[1]	0.5	0.7	0.3	0.4		
	3					1.4					1.1	0.7	1.1[1]	0.3	0.3		
	4					0.4[1]										4.1	3.0
1977	1			1.3	1.0	1.2	1.1						2.3			3.3	2.4
	2			1.6	0.8		0.9				0.3[1]	0.2	1.2	0.2	0.2	1.7	1.4
	3	0.6[1]		1.4	0.9	0.9[1]	1.0				0.8	0.3	1.1	0.2	0.2	1.8	1.6
	4	0.7		1.3	1.0	2.1					1.3	0.7	1.6[1]	0.5	0.5	3.8	2.9
1978	1	0.8		1.2	0.8	2.2					1.0	0.5	1.7[1]	0.4	0.3	2.2[1]	1.6
	2	1.0[1]		1.1	0.7	1.1					0.8	0.4	1.1	0.4	0.3		
	3	0.9		1.4	0.7	1.1					0.8	0.5	1.3	0.4	0.3	1.6	
	4	1.3	1.3	1.6	1.2	3.3					1.7	0.9	1.7	0.5	0.6	1.9	
1979	1	1.0	1.0[1]	1.1	0.7	1.8					0.9	0.4	1.2[1]	0.4	0.4	1.5	
	2	0.4	0.9	1.2	0.6	1.3		0.7	0.9	0.8	0.8	0.4	0.6[1]	0.2	0.3	0.9	
	3	0.6		1.0	0.6	1.6		0.5	0.6	0.8	0.5[1]	0.6[1]	1.1[1]	0.4	0.5	1.0[1]	
	4	0.8[1]		1.2	0.8	1.9					0.7[1]	0.5	0.5[1]	0.3	0.4	0.6[1]	
1980	1	0.9[1]		0.7	0.4		0.3	0.4	0.3	0.3	0.6[1]	0.3	0.3[1]	0.3	0.2	0.7	1.1
	2			0.4	0.4		0.3	0.7	0.4	0.6	0.3[1]	0.3[1]	0.6[1]	0.1	0.2		0.8
	3	0.5		0.7	0.4		0.3	1.0	0.5	0.5		0.2	0.3	0.1	0.1	1.1[1]	1.0
	4	0.6		0.7	0.5		0.4[1]	0.5	0.4	0.4		0.4	0.4	0.3	0.3		1.7
1981	1	0.4		0.5	0.4[1]		0.3	0.2	0.3	0.2	0.7	0.5	0.6	0.3	0.3	1.3	1.0
	2	0.3		0.4	0.3		0.3	0.4	0.3	0.3	0.2	0.2	0.3	0.1	0.2	0.7	0.7
	3			0.4	0.2		0.3	0.3	0.3	0.2	0.5	0.3		0.2	0.3	0.8	0.8
	4			0.4	0.3		0.3[1]	0.4	0.2[1]	0.3	0.8	1.0[1]		0.3	0.4	1.3	1.1

TABLE 7-2. (continued)

		Boston MA	New York NY	Phila. PA		Wash. DC	Detroit MI	Chicago IL			Houston TX		Dallas/Ft.Worth TX			Los Angeles CA	
		Station Type															
Year	Quarter	1	1	1	4	1	1	1	2	3	1	4	1	2	4	1	2
1982	1				0.3	0.5		0.4	0.3	0.3						0.8	0.7
	2		0.5		0.3	0.3		0.2	0.4	0.3						0.5	
	3	1.0	0.5		0.3	0.3		0.3	0.3	0.2		0.2	0.5	0.2	0.2	0.8	
	4		0.6		0.4	0.5[1]		0.4	0.3	0.3		0.3	0.6	0.3	0.2	1.1	0.6
1983	1	0.5	0.3		0.3	0.2		0.4	0.3	0.4	0.4	0.3	1.0	0.6	0.3	1.0	0.6
	2	0.4	0.3		0.2	0.2		0.3	0.3	0.3	0.2			0.2	0.2	0.4	0.3
	3	0.6	0.3		0.2	0.2		0.4	0.2	0.3		0.2	0.6			0.6	0.4
	4	0.5	0.5		0.3	0.3		0.4	0.2	0.2		0.3	0.5			0.8	
1984	1	0.4	0.3		0.3	0.2	0.1	0.3	0.2	0.3		0.4	0.6	0.5	0.2	0.7	0.7
	2	0.4	0.4		0.2	0.2	0.1	0.2	0.3	0.3	0.3	0.1	0.1	0.6	0.5	0.1	0.3
	3	0.5	0.3		0.2		0.1	0.2	0.2	0.3	0.3	0.2	0.4	0.3	0.1		
	4	0.5	0.4		0.3		0.3	0.3	0.3	0.3	0.3	0.1	0.4	0.4	0.2		0.4

Station type:
1. center city commercial
2. center city residential
3. center city industrial
4. suburban residential

[1]: less than required number of 24-hour sampling periods to meet composite criteria.

TABLE 7-3. STATIONS WITH AIR LEAD CONCENTRATIONS GREATER THAN 1.0 μg/m³*

		1979 No. of quarters		Max qtrly ave	1980 No. of quarters		Max qtrly ave	1981 No. of quarters		Max qtrly ave	1982 No. of quarters		Max qtrly ave	1983 No. of quarters		Max qtrly ave	1984 No. of quarters		Max qtrly ave
	Station #	>1.0	>1.5		>1.0	>1.5		>1.0	>1.5		>1.0	>1.5		>1.0	>1.5		>1.0	>1.5	
Birmingham, AL	(028)										1	0	1.32	1	0	1.04			
Leeds, AL	(004)													2	2	3.04	1	1	5.33
" "	(005)													3	2	4.17	2	1	2.96
Troy, AL	(003)	2	2	2.78	2	1	1.89	4	3	3.34	4	4	3.67	3	2	5.44	4	3	7.08
Fairbanks, AK	(010)										1	0	1.01						
Fairbanks, AK	(016)										1	0	1.09						
Glendale, AZ	(001)	1	0	1.06															
Phoenix, AZ	(002A)	1	1	1.54	2	0	1.29	1	0	1.17									
" "	(002G)	2		2.59	2	0	1.49	2	0	1.39									
" "	(004)	2	0	1.48				1	0	1.04									
" "	(013)	2		1.55	1	0	1.06												
" "	(017)										1	0	1.24	1	0	1.08	1	0	1.29
Scottsdale, AZ	(003)	2	0	1.41	1	0	1.13	1	0	1.08									
Tucson, AZ	(009)	1	0	1.18															
Nogales, AZ	(004)				1	0	1.10												
Los Angeles, CA	(001)	1	1	1.51				2	0	1.43									
Anaheim, CA	(001)	1	0	1.11															
Lennox, CA	(001)										2	1	1.68	2	0	1.10			
Los Angeles, CA	(103A)										1	1	1.65						
Los Angeles, CA	(103I)										1	0	1.05						
Lynwood, CA	(001)										1	0	1.25	1	0	1.03	1	0	1.27
Pico Rivera, CA	(001)										1	0	1.15	1	0	1.02			
Adams Co, CO	(001)	2	1	1.77															
Arapahoe Co, CO	(001)	1	0	1.10															
Arvada, CO	(001)	1	1	1.60															
Brighton, CO	(001)	1	0	1.17															
Colorado Springs, CO	(004)	1	0	1.37															
Denver, CO	(001)	2	1	1.70															
" "	(002)	4	3	3.47	2	1	1.53												
" "	(003)	3	1	2.13	1	0	1.03												
" "	(009)	1	1	1.57	2	0	1.23												
" "	(010)	2	1	1.67															
" "	(012)	2	1	1.67	1	0	1.10												

TABLE 7-3. (continued)

	Station #	1979 No. of quarters >1.0	>1.5	Max qtrly ave	1980 No. of quarters >1.0	>1.5	Max qtrly ave	1981 No. of quarters >1.0	>1.5	Max qtrly ave	1982 No. of quarters >1.0	>1.5	Max qtrly ave	1983 No. of quarters >1.0	>1.5	Max qtrly ave	1984 No. of quarters >1.0	>1.5	Max qtrly ave
Englewood, CO	(001)	1	1	1.80															
Garfield, CO	(001)	1	0	1.20															
Grand Junction, CO	(010)	2	1	1.53	1	0	1.27												
Longmont, CO	(001)	2	0	1.07															
Pueblo, CO	(001)	1	0	1.03															
" "	(003)	1	0	1.03															
Routt Co, CO	(003)	1	0	1.33															
New Haven, CT	(123)	3		1.57															
Waterbury, CT	(123)	2	0	1.41															
Wilmington, DE	(002)	2	0	1.21															
Washington, DC	(005)	1	0	1.49															
" "	(007)	4		1.89															
" "	(008)	1	1	1.90															
" "	(011)	2	0	1.44															
" "	(015)	2	0	1.06															
" "	(017)	1	0	1.45															
Dade Co, FL	(020)	1	0	1.16															
Dade Co, FL	(024)																		
Miami, FL	(016)	3	0	1.46	2	0	1.10				1	1	1.51	3	0	1.39			
Perrine, FL	(002)	1	0	1.01															
Hillsborough, FL	(082)	2	0	1.31	1	0	1.09												
Jacksonville, FL	(084)																		
Tampa, FL	(043)	3		1.60	1	0	1.07				3	1	1.72	2	0	1.15	2	0	1.26
Tampa, FL	(060)																		
Boise, ID	(003)				1	0	1.01				1	0	1.10						
Kellogg, ID	(004)	4		9.02	2		6.88												
" "	(006)	4	4	8.25	4	4	8.72	4	4	6.67									
Shoshone Co, ID	(015)	2	0	1.21															
" "	(016)	1	1	2.27	1	0	1.02												
" "	(017)	4		4.57	3		3.33	2	2	1.54									
" "	(020)	2		4.11	2		2.15	1	0	1.49									
" "	(021)	4	4	13.54	4	4	13.67	4	4	11.82									
" "	(027)	4		10.81	3		7.18				2	1	1.75						

TABLE 7-3. (continued)

	Station #	1979 No. of quarters >1.0	 >1.5	Max qtrly ave	1980 No. of quarters >1.0	 >1.5	Max qtrly ave	1981 No. of quarters >1.0	 >1.5	Max qtrly ave	1982 No. of quarters >1.0	 >1.5	Max qtrly ave	1983 No. of quarters >1.0	 >1.5	Max qtrly ave	1984 No. of quarters >1.0	 >1.5	Max qtrly ave
Chicago, IL	(022)				1	0	1.02												
" "	(030)				1	0	1.06												
" "	(005)	1	0	1.05															
" "	(036)	1	0	1.02															
" "	(037)	1	0	1.14															
Cicero, IL	(001)	1	0	1.00															
Elgin, IL	(004)				1	1	1.95												
Granite City, IL	(007)	1	0	1.04															
" "	(009)	4	0	1.15															
" "	(010)	4	4	3.17	3	2	2.97	4	3	7.27							1	0	1.48
" "	(011)	4	0	1.33	1	0	1.43	1	0	1.13									
Jeffersonville, IN	(001)	3	0	1.38															
East Chicago, IN	(001)	2		2.19															
" "	(003)	2	0	1.42															
" "	(004)	1	1	1.67															
" "	(006)	2	0	1.34	1	0	1.04												
Hammond, IN	(004)	2	0	1.18															
" "	(006)	1	0	1.46															
" "	(011)																3	2	2.95
Indianapolis, IN	(030)	1	0	1.16															
Council Bluffs, IA	(017)										1	1	1.59	1	0	1.20			
Des Moines, IA	(051)	1	0	1.30															
Buechel, KY	(001)				1	0	1.41												
Covington, KY	(001)	2	0	1.12															
" "	(008)	1	0	1.16															
Greenup Co, KY	(003)	1	0	1.42															
Jefferson Co, Ky	(029)	1	0	1.05	1	1	1.78												
Louisville, KY	(004)	1	0	1.01	1	1	2.41												
" "	(009)				1	1	1.75												
" "	(019)				1	1	1.59												
" "	(020)				1	1	2.52												
" "	(021)	1	0	1.29	1	1	1.42												
" "	(028)	1	0	1.06															
Newport, KY	(002)	1	0	1.06															
Okolona, KY	(001)	1	1	1.51	2	1	2.31												

TABLE 7-3. (continued)

	Station #	1979 No. of quarters >1.0	 >1.5	Max qtrly ave	1980 No. of quarters >1.0	 >1.5	Max qtrly ave	1981 No. of quarters >1.0	 >1.5	Max qtrly ave	1982 No. of quarters >1.0	 >1.5	Max qtrly ave	1983 No. of quarters >1.0	 >1.5	Max qtrly ave	1984 No. of quarters >1.0	 >1.5	Max qtrly ave
Paducha, KY	(004)	1	0	1.41															
" "	(020)	1	0	1.22															
St. Matthews, KY	(004)	1	0	1.20	1	1	1.83												
Shively, KY	(002)	1	1	1.56															
Baton Rouge, LA	(002)	1	1	1.57															
Portland, ME	(009)	2	0	1.02															
Anne Arundel Co, MD	(001)	1	0	1.27															
" "	(003)	2	0	1.45															
Baltimore, MD	(001)	2	0	1.06															
" "	(006)	1	0	1.09															
" "	(008)	1	0	1.24															
" "	(009)	1	0	1.08															
" "	(018)	2	0	1.12															
Cheverly, MD	(004)	4	1	1.51															
Essex, MD	(001)	2	0	1.15															
Hyattsville, MD	(001)	2	0	1.18															
Springfield, MA	(002)	1	1	1.68	1	0	1.04												
Springfield, MA	(007)										1	0	1.26	1	0	1.00	1	0	1.29
Boston, MA	(002)										2	0	1.08						
Boston, MA	(012)	1	0	1.01															
Eagan, MN	(001)																1	0	1.01
Minneapolis, MN	(027)	1	1	2.44															
" "	(055)				3		2.41	3	1	1.52									
Richfield, MN	(004)	4		1.95	2	0	1.18												
St. Louis Park, MN	(007)	2		2.87	4		3.04												
St. Paul, MN	(031)	1	0	1.04															
" "	(038	1	0	1.36	3		1.82	2	2	3.11	1	1	7.97						
Iron Co, MO	(016)													1	1	2.39	3	2	2.21
" "	(020)													2	1	1.73			
" "	(021)													3	2	2.85	1	0	1.26
" "	(022)													2	1	1.54	1	0	1.10
Jefferson Co, MO	(005)													4	3	4.33	1	0	1.28
" "	(009)													1	0	1.26	1	0	1.11
" "	(011)													4	3	6.70	1	1	2.41
" "	(013)													2	1	2.56	1	1	1.60

TABLE 7-3. (continued)

		1979			1980			1981			1982			1983			1984		
	Station #	No. of quarters >1.0	>1.5	Max qtrly ave	No. of quarters >1.0	>1.5	Max qtrly ave	No. of quarters >1.0	>1.5	Max qtrly ave	No. of quarters >1.0	>1.5	Max qtrly ave	No. of quarters >1.0	>1.5	Max qtrly ave	No. of quarters >1.0	>1.5	Max qtrly ave
Lewis&Clark Co, MT	(002)	4		4.19	4		2.75	2	2	3.19	2	2	2.25	4	4	3.12	4	4	3.87
Lewis&Clark Co, MT	(007)													2	2	5.26	4	4	6.83
" "	(008)				1	0	1.19							2	0	1.31	3	0	1.48
" "	(714)													1	1	1.99			
" "	(716)													4	4	3.39			
Lewis&Clark Co, MT	(719)										2	2	2.69	2	2	1.84	4	3	4.63
Lewis&Clark Co, MT	(722)										2	2	2.34	3	2	2.96	3	4	3.23
" "	(724)													1	0	1.23			
Omaha, NE	(011)										1	0	1.21						
Omaha, NE	(034)	1	0	1.08															
Las Vegas, NV	(001)	1	0	1.15															
Clifton, NJ	(002)										1	0	1.17						
Newark, NJ	(001)	1	0	1.17															
New Brunswick, NJ	(003)																1	1	1.73
Perth Amboy, NJ	(001)	1	0	1.08															
Paterson, NJ	(001)	1	0	1.42															
Elizabeth, NJ	(002)	1	0	1.16															
Salem Co, NJ	(003)										1	0	1.24	2	1	1.81			
Albuquerque, NM	(022)										1	0	1.01						
Dona Ana Co, NM	(015)										3	0	1.34						
Orange Co, NY	(001)										1	0	1.03						
Yonkers, NY	(001)	1	0	1.08															
Cincinnati, OH	(001)	1	0	1.15															
Portland, OR	(082)										4	2	1.63	4	0	1.37	3	1	1.58
Laureldale, PA	(717)	4		3.30	2		1.86	4	3	2.18	3	0	1.49	4	0	1.25	1	0	1.40
Reading, PA	(712)	1	0	1.11															
E.Conemaugh, PA	(804)	3	0	1.28															
Throop, PA	(019)	3	0	1.13															
Lancaster City, PA	(315)	1	0	1.18															
New Castle, PA	(015)	1	0	1.01															
Montgomery Co, PA	(103)	1	0	1.23															
Pottstown, PA	(101)	1	0	1.16															
Phila., PA	(026)	3	0	1.21															
" "	(028)	4		2.71	3	0	1.26	1	0	1.30	3	1	1.57	3	2	3.66	4	4	5.13

TABLE 7-3. (continued)

	Station #	1979 No. of quarters >1.0	1979 No. of quarters >1.5	Max qtrly ave	1980 No. of quarters >1.0	1980 No. of quarters >1.5	Max qtrly ave	1981 No. of quarters >1.0	1981 No. of quarters >1.5	Max qtrly ave	1982 No. of quarters >1.0	1982 No. of quarters >1.5	Max qtrly ave	1983 No. of quarters >1.0	1983 No. of quarters >1.5	Max qtrly ave	1984 No. of quarters >1.0	1984 No. of quarters >1.5	Max qtrly ave
" "	(031)	2	0	1.29															
" "	(038)	1	0	1.06															
Guaynabo Co, PR	(001)	2		1.60	1	0	1.06	1	0	1.02									
Ponce, PR	(002)	1	0	1.08															
San Juan Co., PR	(003)	4		3.59							1	1	1.69	4	0	1.27	4	0	1.30
E.Providence, RI	(008)	2	0	1.10															
Providence, RI	(007)	4		1.92	2	0	1.16				2	0	1.11						
" "	(015)	1	0	1.34															
Greenville, SC	(001)	2	0	1.38															
Memphis, TN	(035)										1	0	1.30	1	0	1.17	3	1	6.19
Nashville/Davidson, TN	(006)	1	0	1.05															
San Antonio, TX	(034)	1	0	1.23															
Dallas, TX	(018)	1	1	1.59															
" "	(029)	1	0	1.07															
" "	(035)	1	0	1.12															
" "	(046)	1	0	1.22															
" "	(049)	1	0	1.01															
" "	(050)	2	0	1.13															
" "	(057)													1	0	1.23			
" "	(060)													1	0	1.44	3	1	1.52
El Paso, TX	(002A)	1	1	1.90			2.12												
" "	(002F)	1	1	1.90				4	1	1.79				2	1	1.37			
" "	(002G)	4		2.60													1	0	1.39
" "	(018)	2		1.91															
" "	(021)	1	0	1.02															
" "	(022)	2		1.84															
" "	(023)	2		2.12															
" "	(027)	2		2.15	2		1.74	4	2	1.75				3	1	1.54	3	1	1.54
" "	(028)				1	0	1.16												
" "	(030)	1	0	1.02															
" "	(031)	1	1	2.47															
" "	(033)	1	1	1.97													1	0	1.02
" "	(037)																		

TABLE 7-3. (continued)

	Station #	1979 No. of quarters >1.0	 >1.5	Max qtrly ave	1980 No. of quarters >1.0	 >1.5	Max qtrly ave	1981 No. of quarters >1.0	 >1.5	Max qtrly ave	1982 No. of quarters >1.0	 >1.5	Max qtrly ave	1983 No. of quarters >1.0	 >1.5	Max qtrly ave	1984 No. of quarters >1.0	 >1.5	Max qtrly ave
Houston, TX	(001)	2	0	1.35															
" "	(002)	2	0	1.39															
" "	(037)	1	0	1.26															
" "	(049)	3	0	1.13															
Ft. Worth, TX	(003)	2	0	1.14				1	1	1.96							3	1	1.60
Seattle, WA	(057)	1	0	1.36															
Tacoma, WA	(004)	1	0	1.06										1	0	1.04			
Charleston, WV	(001)	1	0	1.09															

*Where data are not given, reported quarterly averages were less than 1.0 μg/m³ or there were insufficient reports for valid quarterly averages.

near stationary point sources such as smelters, or near routes of high traffic density. Both situations are represented in Table 7-3; e.g., the Idaho data reflect predominantly stationary source emissions, whereas the Washington, D.C. data reflect predominantly vehicular emissions.

Table 7-4 summarizes the maximum quarterly lead values for those stations reporting 4 valid quarters in 1979, 1980, and 1981, grouped according to principal exposure orientation or influence--population, stationary source, or background. The sites located near stationary sources clearly dominate the concentrations over 2.0 μg/m³; however, new monitor siting guidelines, discussed in Section 7.2.1.3.2, will probably effect some increase in the upper end of the distribution of values from population-oriented sites by adding monitoring sites closer to traffic emissions.

The effect of the 1978 National Ambient Air Quality Standard for Lead has been to reduce the air concentration of lead in major urban areas. Similar trends may also be seen in urban areas of lesser population density. Continuous monitoring at non-urban stations has been insufficient to show a trend at more than a few locations. There are two reports that reflect a trend toward decreasing atmospheric lead concentrations. Eisenreich et al. (1986) report decreasing concentrations of lead in rain during the period 1979-83 from 29 to 4.3 μg/liter in urban areas and 5.7 to 1.5 in rural areas. All sites were in Minnesota. Trefry et al. (1985) reported a decrease in the lead concentration of Mississippi River sediment layers for the post 1970 period. They estimated that the Mississippi River carried 40 percent less lead in suspended sediments in 1982-83 than in 1974-1975.

7.2.1.3 Changes in Air Lead Prior to Human Uptake. There are many factors that can cause differences between the concentration of lead measured at a monitoring station and the actual inhalation of air by humans. The following sections show that air lead concentrations usually decrease with vertical and horizontal distance from emission sources, and are generally lower indoors than outdoors. A person working on the fifth floor of an office building would be exposed to less lead than a person standing on a curb at street level. The following discussions will describe how these differences can affect individual exposures in particular circumstances.

7.2.1.3.1 Airborne particle size distributions. The effects of airborne lead on human health and welfare depend upon the sizes of the lead-containing particles. As discussed in Chapter 6, large particles are removed relatively quickly from the atmosphere by dry and wet deposition processes. Particles with diameter smaller than a few micrometers tend to remain airborne for long periods (see Section 6.3.1).

Figure 7-5 summarizes airborne lead particle size data from the literature (Davidson and Osborn, 1984). Minimum and maximum aerodynamic particle diameters of 0.05 μm and 25 μm, respectively, have been assumed unless otherwise specified in the original reference. Note that most of the airborne lead mass is associated with small particles. There is also a distinct

TABLE 7-4. DISTRIBUTION OF AIR LEAD CONCENTRATIONS BY TYPE OF SITE FOR 1980-83

Category	Concentration ranges ($\mu g/m^3$) ≦0.5	>0.5 ≦1.0	>1.0 ≦1.5	>1.5 ≦2.0	>2.0	Total no. of site-years
Neighborhood scale	38	20	3	0	1	62
Middle scale	13	14	6	0	0	33
Stationary source	99	25	13	5	17	159
Microscale roadside	5	12	8	4	1	30
Other[1]	666	190	30	15	4	905
Total	820	262	59	24	24	1189
Percentage of sites in concentration range	69%	22%	5%	2%	2%	100%

[1] Data are the number of site years during 1980-83 falling within the designated quarterly average concentration range. To be included, a site year must have four valid quarters of data.

Source: SAROAD system.

peak of large particles in the upper end of many of the distributions. Two separate categories of sources are responsible for these distributions: the small particles result from nucleation of vapor phase lead emissions (predominantly automotive), while the larger particles may originate directly from soil dust, coal particles, and other coarse materials, or indirectly by the attachment of anthropogenically emitted submicron particles with high lead content to larger particles, such as soil particles. Large particle peaks may also indicate fly ash with a surface coating of condensed lead (Linton et al., 1980).

Information associated with each in the distributions in Figure 7-5 may be found in Table 7A-1 of Appendix 7A. The first six distributions were obtained by an EPA cascade impactor network established in several cities during the calendar year 1970 (Lee et al., 1972). These distributions represent the most extensive size distribution data base available. However, the impactors were operated at excessive air flow rates that most likely resulted in particle bounceoff, biasing the data toward smaller particles (Dzubay et al., 1976). Many of the later distributions, although obtained by independent investigators with poorly defined quality control, were collected using techniques that minimize particle bounceoff and hence may be more reliable. It is important to note that a few of the distributions were obtained without backup filters that capture the smallest particles. These distributions are likely to be inaccurate, since an appreciable fraction of the airborne lead mass was probably not sampled. The distributions of Figure 7-5 have been used with published lung deposition data to estimate the

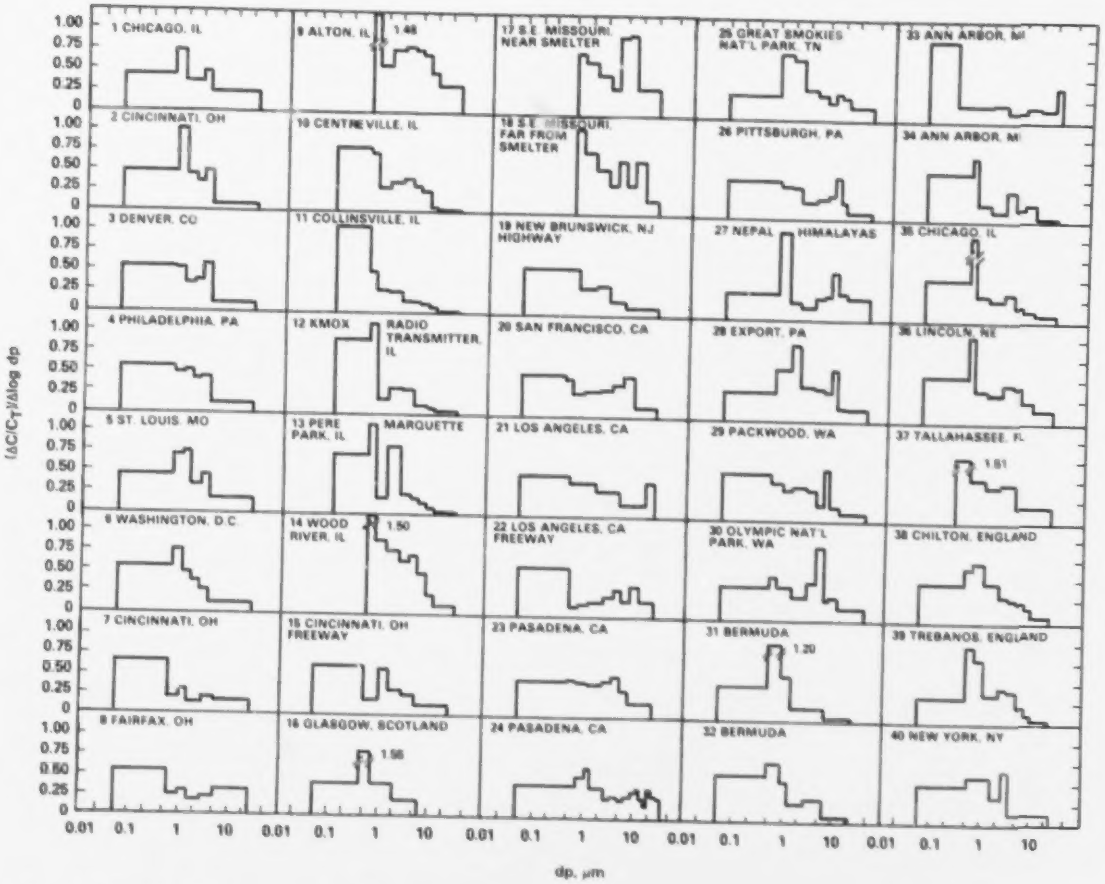

Figure 7-5. Airborne mass size distributions for lead taken from the literature. ΔC represents the airborne lead concentration in each size range. C_T is the total airborne lead concentration in all size ranges, and d_p is the aerodynamic particle diameter.

Source: Davidson and Osborn (1984).

fraction of inhaled airborne lead deposited in the human respiratory system (see Section 10.2.1).

7.2.1.3.2 Vertical gradients and siting guidelines. New guidelines for placing ambient air lead monitors went into effect in July, 1981 (C.F.R. (1984) 40: §58, see Section 4.2.1). "Microscale" sites, placed between 5 and 15 meters from thoroughfares and 2 to 7 m above the ground, are prescribed, but until now few monitors have been located close to heavily traveled roadways. Many of these microscale sites might be expected to show higher lead concentrations than that measured at nearby middlescale urban sites, due to vertical gradients in lead concentrations near the source. One study (PEDCo, 1981) gives limited insight into the relationship between a microscale location and locations further from a roadway. The data in the lower half of Table 7-5 summarize total suspended particulates and particulate lead concentrations in samples collected in Cincinnati, Ohio, on 21 consecutive days in April and May, 1980, adjacent to a 58,500 average daily traffic (ADT) expressway connector. Simple interpolation indicates that a microscale monitor as close as 5 meters from the roadway and 2 m above the ground would record concentrations some 20 percent higher than those at a "middle scale" site 21.4 m from the roadway. On the other hand, these data also indicate that although lead concentrations very close to the roadway (2.8 m setback) are quite dependent on the height of the sampler, the averages at the three selected heights converge rapidly with increasing distance from the roadway. In fact, the average lead concentration (1.07 $\mu g/m^3$) for the one monitor (6.3 m height, 7.1 m setback) that satisfies the microscale site definition does not prove to be significantly different from the averages for its two companions at other heights but the same 7.1 m setback, or from the averages for any of the three monitors at the 21.4 m setback. It also appears that distance from the source, whether vertical or horizontal, can be the primary determining factor for changes in air lead concentrations. At 7.1 m setback distance, the samplers at heights of 1.1 and 6.3 m would be about 7 and 11 m, respectively, from the road surface. The values at these vertical distances are only slightly lower than the corresponding values for comparable horizontal distances.

Other urban locations around the country with their own characteristic wind flow patterns and complex settings, such as multiple roadways, may produce situations where the microscale site does not record the highest concentrations. Collectively, however, the addition of these microscale sites to the nation's networks can be expected to shift the distribution of reported quarterly averages toward higher values. This shift will result from the change in composition of the networks and is a separate phenomenon from downward trend at long-established sites described above, reflecting the decrease in lead additives used in gasoline.

Two other studies show that lead concentrations decrease with vertical distance from the source (PEDCo Environmental, 1977; Sinn, 1980). PEDCo Environmental (1977) measured lead concentrations at heights of 1.5 and 6.1 m at sites in Kansas City, MO and Cincinnati, OH (top

TABLE 7-5. VERTICAL DISTRIBUTION OF LEAD CONCENTRATIONS

Location	Setback distance (m)	Height (m)	Effective[a] distance from source (m)	Air lead conc. (μg/m³)	Ratio to source
Kansas City[b]					
east of road	3.0[d]	6.1	6.4	1.7	0.85
		1.5	3.2	2.0	S[e]
west of road	3.0[d]	6.1	6.4	1.5	0.88
		1.5	3.2	1.7	S
Cincinnati[b]					
east of road	3.0[d]	6.1	6.4	0.9	0.64
		1.5	3.2	1.4	S
west of road	3.0[d]	6.1	6.4	0.6	0.75
		1.5	3.2	0.8	S
Cincinnati[c]	2.8	10.5	10.4	0.81	0.61
		6.3	6.4	0.96	0.72
		1.1	2.9	1.33	S
Cincinnati[c]	7.1	10.5	12.3	0.93	0.69
		6.3	9.2	1.07	0.80
		1.1	7.1	1.16	0.87
Cincinnati[c]	21.4	10.5	23.6	0.90	0.68
		6.3	22.2	0.97	0.73
		1.1	21.4	1.01	0.77

[a] Effective distance was calculated assuming the source was the edge of the roadway at a height of 0.1 m.

[b] Source: PEDCo Environmental (1977).

[c] Source: PEDCo (1981).

[d] Assumed setback distance of 3.0 m.

[e] Station closest to source used to calculate ratio.

half of Table 7-5). The sampling sites in Kansas City were described as unsheltered, unbiased by local pollution influences, and not immediately surrounded by large buildings. The Cincinnati study was conducted in a primarily residential area with one commercial street. Samplers were operated for 24-hour periods; however, a few 12-hour samples were collected from 8 AM to 8 PM. Data were obtained in Kansas City on 35 days and in Cincinnati on 33 days. The measured concentrations were greater at 1.5 m than at 6.1 m, and the difference between the east side and west side of the street was approximately the same as the difference between 1.5 m and 6.1 m in height.

Sinn (1980) investigated airborne lead concentrations at heights of 3 and 20 m above a road in Frankfurt, Germany. Measurements conducted in December 1975, December 1976, and January 1978 gave monthly mean values of 3.18, 1.04, and 0.66 μg/m^3, respectively, at 3 m height. The corresponding values at 20 m height were 0.59, 0.38, and 0.31 μg/m^3, showing a substantial reduction at this height. The decrease in concentration over the 2-year period was attributed to a decrease in the permissible lead content of gasoline from 0.4 to 0.15 g/liter beginning in January, 1976.

Two reports show no relationship between air concentration and vertical distance (Barltrop and Strehlow, 1976; Ter Haar, 1979). From August, 1975 to July, 1976, Barltrop and Strehlow (1976) conducted an air sampling program in London at a proposed nursery site under an elevated motorway. The height of the motorway was 9.3 m. Air samplers were operated at five to seven sites during the period from Monday to Friday, 8 AM to 6 PM, for one year. The maximum individual value observed was 18 μg/m^3. The 12-month mean ranged from 1.35 to 1.51 μg/m^3, with standard deviations of 0.91 and 0.66, respectively. The authors reported that the airborne concentrations were independent of height from ground level up to 7 m.

Ter Haar (1979) measured airborne lead at several heights above the ground, using samplers positioned 6 m from a heavily traveled road in Detroit. A total of nine 8-hour daytime samples were collected. The overall average airborne lead concentrations at heights of 0.3, 0.9, 1.5, and 3.0 m were 4.2, 4.8, 4.7, and 4.6 μg/m^3, respectively, indicating a uniform concentration over this range of heights at the measurement site. It should be noted that at any one height, the concentration varied by as much as a factor of 10 from one day to the next; the importance of simultaneous sampling when attempting to measure gradients is clearly demonstrated.

Data that show variations with vertical distance reflect the strong influence of the geometry of the boundary layer, wind, and atmospheric stability conditions on the vertical gradient of lead resulting from automobile emissions. The variability of concentration with height is further complicated by the higher emission elevation of smokestacks. Concentrations measured from sampling stations on the roofs of buildings several stories high may not reflect actual human exposure conditions, but neither would a single sampling station located at ground level among several buildings. The height variation in concentration resulting from vertical diffusion of automobile emissions is likely to be small compared to temporal and spatial variations resulting from surface geometry, wind, and atmospheric conditions. Our understanding of the complex factors affecting the vertical distribution of airborne lead is extremely limited, but the data of Table 7-5 indicate that air lead concentrations are primarily a function of distance from the source, whether vertical or horizontal.

7.2.1.3.3 Indoor/outdoor relationships and personal monitoring. Because people spend much of their time indoors, ambient air sampled outdoors may not accurately represent actual inhalation exposure to airborne lead. Table 7-6 summarizes the results of several indoor/outdoor airborne lead studies. In nearly all cases, the indoor concentration is substantially lower than the corresponding value outdoors; the only indoor/outdoor ratio exceeding unity is for a high-rise apartment building, where air taken in near street level is rapidly distributed through the building air circulation system. Some of the studies in Table 7-6 show smaller indoor/outdoor ratios during the winter, when windows and doors are tightly closed. Overall, the data suggest indoor/outdoor ratios of 0.6-0.8 are typical for airborne lead in houses without air conditioning. Ratios in air conditioned houses are expected to be in the range of 0.3-0.5 (Yocom, 1982). The available data imply that virtually all airborne lead found indoors is associated with material transported from the outside. Because of the complexity of factors affecting infiltration of air into buildings, however, it is difficult to predict accurately indoor lead concentrations based on outdoor levels. Rabinowitz et al. (1984) found a correlation between indoor air lead in Boston homes and the amount of lead sold in gasoline in Massachusetts.

Even detailed knowledge of indoor and outdoor airborne lead concentrations at fixed locations may still be insufficient to assess human exposure to airborne lead. The study of Tosteson et al. (1982) in Table 7-6 included measurement of airborne lead concentrations using personal exposure monitors carried by individuals going about their day-to-day activities. In contrast to the lead concentrations of 0.092 and 0.12 μg/m^3 at fixed locations, the average personal exposure was 0.16 μg/m^3. The authors suggest this indicates an inadequacy of using fixed monitors at either indoor or outdoor locations to assess exposure.

Rohbock (1981) reported that, whereas a parked car may exhibit properties similar to buildings in reducing internal air concentrations, a moving car quickly reaches the same air lead concentration inside as outside, suggesting a rapid exchange of air in a moving vehicle.

7.2.2 Lead in Soil

Much of the lead in the atmosphere is transferred to terrestrial surfaces where it is eventually passed to the upper layer of the soil surface. The mechanisms that determine the transfer rate of lead to soil are described in Section 6.4.1 and the transformation of lead in soil in Section 6.5.1. The uptake of lead by plants and its subsequent effect on animals may be found in Sections 8.3 and 8.4, respectively. The purpose of this section is to discuss the distribution of lead in U.S. soils and the impact of this lead on potential human exposures.

TABLE 7-6. COMPARISON OF INDOOR AND OUTDOOR AIRBORNE LEAD CONCENTRATIONS

Type of building	Airborne lead concentration ($\mu g/m^3$)		Indoor/outdoor ratio	Location	Ref
	Indoor	Outdoor			
Library	1.12	2.44	0.46	Hartford, CT	1
City hall	1.31	1.87	0.70	"	
Office building 1	0.73	1.44	0.51	"	
Office building 2	0.55	1.09	0.51	"	
House 1	1.37	2.48	0.55	"	
House 2	0.94	1.34	0.70	"	
Apartment building 1					
Second floor	1.46	2.67	0.55	New York, NY	2
Roof	1.50	1.38	1.09	"	
Apartment building 2					
Third floor	--	1.21	--	"	
Eleventh floor	1.68	--	--	"	
Eighteenth floor	1.86	--	--	"	
Roof	--	1.42	--	"	
New air conditioned apartment	0.12-0.40	0.13-0.50	0.82	New York, NY	3
Older non-air conditioned apartment	0.14-0.51	0.17-0.64	0.87	"	
Air conditioned public building	0.15-0.79	0.33-1.18	0.63	"	
Non-air conditioned storeroom in public building	0.45-0.98	0.38-1.05	0.81	"	
Houses	--	--	0.53	Pittsburgh, PA	4
University buildings	--	--	0.28	"	
Public schools	--	--	0.28	"	
Store	--	--	0.31	"	
Commercial office	--	--	0.27	"	
Houses	0.092	0.12	0.74	Topeka, KS	5
Houses with gas stoves	--	--	0.65	Boston, MA	6
Houses with electric stoves	--	--	0.68	"	
Office buildings	--	--	0.42	"	

TABLE 7-6. (continued)

Type of building	Airborne lead concentration (μg/m³) Indoor	Outdoor	Indoor/outdoor ratio	Location	Ref
House 1					
Before energy conservation retrofit	0.039	0.070	0.56	Medford, OR	7
After energy conservation retrofit	0.037	0.084	0.44		"
House 2					
Before energy conservation retrofit	0.035	0.045	0.78	"	
After energy conservation retrofit	0.038	0.112	0.34	"	

Sources:

1. Yocom et al., 1971.
2. General Electric Company, 1972.
3. Halpern, 1978.
4. Cohen and Cohen, 1980.
5. Tosteson et al., 1982.
6. Geomet, Inc., 1981.
7. Berk et al., 1981.

7.2.2.1. Typical Concentrations of Lead in Soil

7.2.2.1.1 Lead in urban, smelter, and rural soils. Shacklette et al. (1971) sampled soils at a depth of 20 cm to determine the elemental composition of soil materials derived from the earth's crust, not the atmosphere. The range of values probably represent natural levels of lead in soil, although there may have been some contamination with anthropogenic lead during collection and handling. Lead concentrations in soil ranged from less than 10 to greater than 70 μg/g. The arithmetic mean of 20 μg/g and geometric mean of 16 μg/g reflect the fact that most of the 863 samples were below 30 μg/g at this depth. McKeague and Wolynetz (1980) found the same arithmetic mean (20 μg/g) for 53 uncultivated Canadian soils. The range was 5 to 50 μg/g and there was little variation with depth between the A, B and C horizons in the soil profile.

Studies discussed in Section 6.5.1 have determined that atmospheric lead is retained in the upper 2-5 cm of undisturbed soil, especially soils with at least 5 percent organic matter and a pH of 5 or above. There has been no general survey of this upper 2-5 cm of the soil surface in the United States, but several studies of lead in soil near roadsides and smelters and a few studies of lead in soil near old houses with lead-based paint can provide the backgound information for determining potential human exposures to lead from soil.

Because lead is immobilized by the organic component of soil (Section 6.5.1), the concentration of anthropogenic lead in the upper 2-5 cm is determined by the flux of atmospheric lead to the soil surface. Near roadsides, this flux is largely by dry deposition and the rate depends on particle size and concentration. These factors vary with air concentration and average windspeed (see Section 6.4.1). In general, deposition drops off abruptly with increasing distance from the roadway. This effect is demonstrated in studies that show that surface soil lead decreases exponentially up to 25 m from the edge of the road. The original work of Quarles et al. (1974) showed decreases in soil lead from 550 to 40 µg/g within 25 m alongside a highway with 12,500 vehicles/day in Virginia. Pierson and Brachaczek (1976) found that lead concentrations in topsoil adjacent to a major artery decreased exponentially from 0 to 12 m away from the highway (Figure 7-6). These findings were confirmed by Wheeler and Rolfe (1979), who observed an exponential decrease linearly correlated with traffic volume. Agrawal et al. (1981) found similar correlations between traffic density and roadside proximity in Baroda City, as did Garcia-Miragaya et al. (1981) in Venezuela and Wong and Tam (1978) in Hong Kong. Little and Wiffen (1978) found additional relationships between particle size and roadside proximity and decreases with depth in the soil profile. The general conclusion from these studies is that roadside soils may contain atmospheric lead from 30-2000 µg/g in excess of natural levels within 25 m of the roadbed, all of which is in the upper layer of the soil profile. It is assumed that particles deposited directly on the roadway are washed to the edge of the pavement, but do not migrate beyond the shoulder.

Near primary and secondary smelters, lead in soil decreases exponentially within a 5-10 km zone around the smelter complex. Soil lead contamination varies with the smelter emission rate, stack height, length of time the smelter has been in operation, prevailing windspeed and direction, regional climatic conditions, and local topography (Roberts, 1975).

Little and Martin (1972) observed decreases from 125 to 10 µg/g in a 6 km zone around a smelting complex in Great Britain; all of the excess lead was in the upper 6 cm of the soil profile. Roberts (1975) reported soil lead between 15,000 and 20,000 µg/g near a smelter in Toronto. Kerin (1975) found 5,000-9,000 µg/g adjacent to a Yugoslavian smelter; the contamination zone was 7 km in radius. Ragaini et al. (1977) observed 7900 µg/g near a smelter in Kellogg, Idaho; they also observed a 100-fold decrease at a depth of 20 cm in the soil profile. Palmer and Kucera (1980) observed soil lead in excess of 60,000 µg/g near two smelters in Missouri, decreasing to 10 µg/g at 10 km.

Urban soils may be contaminated from a variety of atmospheric and non-atmospheric sources. The major sources of soil lead seem to be paint chips from older houses and deposition from nearby highways. Lead in soil adjacent to a house decreases with distance from the

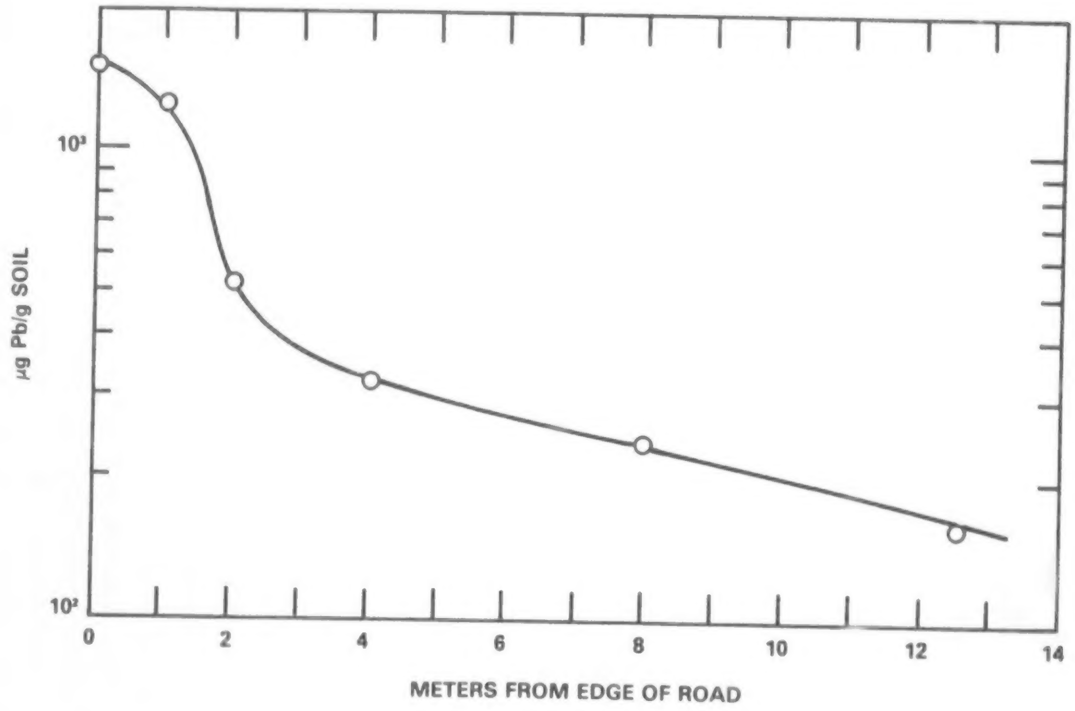

Figure 7-6. Change in soil lead concentrations with increasing distance from a roadway.

Source: Pierson and Brachaczek (1976).

house; this may be due to paint chips or to dust of atmospheric origin washing from the rooftop (Wheeler and Rolfe, 1979).

Andresen et al. (1980) reported lead in the litter layer of 51 forest soils in the northeastern United States. They found values from 20-700 µg/g, which can be compared only qualitatively to the soil lead concentration cited above. This study clearly shows that the major pathway of lead to the soil is by the decomposition of plant material containing high concentrations of atmospheric lead on or within their tissues. Because this organic matter is a part of the decomposer food chain, and because the organic matter is in dynamic equilibrium with soil moisture, it is reasonable to assume that lead associated with organic matter is biologically more mobile than lead tightly bound within the crystalline structure of inorganic rock fragments.

Finally, a definitive study that describes the source of soil lead was reported by Gulson et al. (1981) for soils in the vicinity of Adelaide, South Australia. In an urban to rural transect, stable lead isotopes were measured in the top 10 cm of soils over a 50 km distance. By their isotopic compositions, three sources of lead were identified: natural, non-automotive industrial lead from Australia, and tetraethyl lead manufactured in the United States. The results indicated that most of the soil surface lead originated from leaded gasoline. Similar studies have not been conducted in the United States.

7.2.2.1.2 Natural and anthropogenic sources of soil lead. Although no study has clearly identified the relative concentrations of natural and anthropogenic lead in soil, a few clarifying statements can be made with some certainty. Lead may be found in inorganic primary minerals, on humic substances, complexed with Fe-Mn oxide films, on secondary minerals or in soil moisture. All of the lead in primary minerals is natural and is bound tightly within the crystalline structure of the minerals. Most of this lead can be released only by harsh treatment with acids. The lead on the surface of these minerals is leached slowly into the soil moisture. Atmospheric lead forms complexes with humic substances or on oxide films that are in equilibrium with soil moisture, although the equilibrium strongly favors the complexing agents. Consequently, the ratio of anthropogenic to natural lead in soil moisture depends mostly on the amounts of each type of lead in the complexing agents and very little on the concentration of natural lead in the inorganic minerals.

Except near roadsides and smelters, only a few micrograms of atmospheric lead have been added to each square centimeter of soil surface. Several studies indicate that this lead is available to plants (Section 8.3.1.1). Even with small amounts of atmospheric lead, as much as 75 percent of the lead in soil moisture is of atmospheric origin (Elias et al., 1982). A conservative estimate of 50 percent is used in the discussions in Section 7.3.1.2. A breakdown of the types of lead in soil may be found in Table 7-7.

TABLE 7-7. SUMMARY OF SOIL LEAD CONCENTRATIONS
(μg/g)

Matrix	Natural lead	Atmospheric lead		Total lead	
		Rural	Urban	Rural	Urban
Total soil	8-25	3-5	50-150	10-30	150-300
Primary minerals	8-25	-	-	8-25	8-25
Humic substances*	20	60	2000	80	2000
Soil moisture	0.0005	0.0005	0.0150	0.001	0.0155

*Assumes 5% organic matter, pH 5.0; may also include lead in Fe-Mn oxide films.

7.2.2.2 Pathways of Soil Lead to Human Consumption

7.2.2.2.1 Crops. On the surfaces of vegetation, most lead may be of atmospheric origin. In the internal tissues, lead may be a combination of atmospheric and soil origin. As with soils, lead on vegetation surfaces decreases exponentially with distance away from roadsides and smelters (Cannon and Bowles, 1962; Nasralla and Ali, 1985; see also Chapter 8). For many years, plant surfaces have been used as indicators of lead pollution (Garty and Fuchs, 1982; Pilegaard, 1978; Ratcliffe, 1975; Ruhling and Tyler, 1969; Tanaka and Ichikuni, 1982). These studies all show that lead on the surface of leaves and bark is proportional to traffic density and distance from the highway, or more specifically, to air lead concentrations and particle size distributions. Other factors such as surface roughness, wind direction and speed are discussed in Chapter 6. The data also show that lead in internal plant tissues is directly, although not always linearly, related to lead in soil. Nicklow et al. (1983) found a linear relationship between extracted soil lead and several food crops.

In a study to determine the background concentrations of lead and other metals in agricultural crops, the Food and Drug Administration (Wolnik et al., 1983, 1985), in cooperation with the U.S. Department of Agriculture and the U.S. Environmental Protection Agency, analyzed over 1500 samples of the most common crops taken from a cross section of geographic locations. Collection sites were remote from mobile or stationary sources of lead. Soil lead concentrations were within the normal range (8-25 μg/g) of U.S. soils. Extreme care was taken to avoid contamination during collection, transportation, and analysis. The concentrations of lead in crops found by Wolnik et al. (1983, 1985) are shown as "Total" concentrations in Table 7-8. The breakdown by source of lead is discussed below. The total concentration data should probably be seen as representing the lowest concentrations of lead in food available to Americans. From harvest to packaging, the lead concentration in food increases by a factor of 2-12 (see Section 7.3.1.2). A small portion of this increase may occur because: (1) some

TABLE 7-8. BACKGROUND LEAD IN BASIC FOOD CROPS AND MEATS
(μg/g fresh weight)

Crop	Natural Pb	Indirect atmospheric	Direct atmospheric	Total[†]
Wheat	0.0015	0.0015	0.034	0.037
Potatoes	0.0045	0.0045	--	0.009
Field corn	0.0015	0.0015	0.019	0.022
Sweet corn	0.0015	0.0015	--	0.003
Soybeans	0.021	0.021	--	0.042
Peanuts	0.005	0.005	--	0.010
Onions	0.0023	0.0023	--	0.0046
Rice	0.0015	0.0015	0.004	0.007
Carrots	0.0045	0.0045	--	0.009
Tomatoes	0.001	0.001	--	0.002
Spinach	0.0015	0.0015	0.042	0.045
Lettuce	0.0015	0.0015	0.010	0.013
Beef (muscle)	0.0002	0.002	0.02	0.02[*]
Pork (muscle)	0.0002	0.002	0.06	0.06[*]

[†] Except as indicated, data are from Wolnik et al. (1983, 1985).

[*] Data from Penumarthy et al. (1980).

crops are grown closer to highways and stationary sources of lead than those sampled by Wolnik et al. (1983, 1985); (2) some harvest techniques used by farmers might add more lead to the crop than did Wolnik et al.; and (3) some crops are grown on soils significantly higher in lead than those of the Wolnik et al. study because of a history of fertilizer additions or sludge applications.

Because the study reported by Wolnik et al. was a systematic effort that covers a broad spectrum of agricultural practices in the United States and was conducted with appropriate attention to quality assurance, it serves in this report as the sole basis for background crop data. There are many other reputable studies that describe the impact of lead on crops under specific circumstances or with a variety of control measures. Generally, these studies report that the lead concentrations are highest in leafy crops, lowest in fruits, with root crops somewhat intermediate (e.g., Nicklow et al., 1983). It is important to recognize that root crops such as radishes and potatoes are specialized structures for the storage of photosynthetic products, and are functionally different from the roots that absorb water and nutrients. These latter roots usually have lead concentrations higher than shoots or leaves and form a reasonably effective barrier to soil lead. Reports of lead in food crops from other countries have found patterns similar to those in the United States (Nasralla and Ali, 1985; Wong and Koh, 1982).

Studies that specifically apply to roadside or stationary source conditions can be evaluated in the context of these recent background findings by Wolnik et al. (1983, 1985). Studies of the lead associated with crops near highways have shown that both lead taken up from soil and aerosol lead delivered by deposition are found associated with the edible portions of common vegetable crops. However, there is enormous variability in the total amount of lead associated with such crops and in the relative amounts of lead in the plants versus on the plants. The variability depends upon several factors, the most prominent of which are the plant species, the traffic density, the meteorological conditions, and the local soil conditions (Welch and Dick, 1975; Rabinowitz, 1974; Dedolph et al., 1970; Motto et al., 1970; Schuck and Locke, 1970; Ter Haar, 1970). These factors, coupled with the fact that many studies have not differentiated between lead on plants versus lead in plants, make it difficult to generalize on the relationship between lead in crops and lead in soil or air. Data of Schuck and Locke (1970) suggest that in some cases (e.g., tomatoes and oranges) much of the surface lead is readily removed by washing. But as noted in Section 6.4.3, this is not universally true; in some cases, much more vigorous washing procedures would be necessary to remove all or most of the surface lead.

Ter Haar (1970) found that inedible portions of several plants (bean leaves, corn husks, soybean husks, and chaff from oats, wheat, and rice) had two to three times the lead concentration when grown near a busy highway compared with similar plants grown 160 m from the highway or in a greenhouse supplied with filtered air. The edible portions of these and other plants showed little or no difference in lead content between those grown in ambient air and those grown in the filtered air. However, the lead concentrations found by Ter Haar (1970) for edible portions of crops grown in filtered air in the greenhouse were generally one to two orders of magnitude higher than those of the same types of crops taken from actual agricultural situations by Wolnik et al. (1983, 1985). Dedolph et al. (1970) found that while ryegrass and radish leaves grown near a busy highway contained deposited airborne lead, the edible portion of the radish was unaffected by variations in either soil lead or air lead.

The accumulation of lead by edible portions of crops was measured by Ter Haar (1970), who showed that edible plant parts not exposed to air (potatoes, corn, carrots, etc.) do not accumulate atmospheric lead, while leafy vegetables do. These results were confirmed by McLean and Shields (1977), who found that most of the lead associated with food crops is on leaves and husks. The general conclusion from these studies is that lead associated with food crops varies according to exposure to the atmosphere and in proportion to the effort taken to separate husks, chaff, and hulls from edible parts during processing for human or animal consumption.

To estimate the distribution of natural and atmospheric lead in food crops (Table 7-8), it is necessary to recognize that some crops of the Wolnik et al. study have no lead from direct atmospheric deposition, but rather that all lead found in these crops comes through soil moisture. The lowest concentrations of lead are found in those crops where the edible portion grows above ground and it does not accumulate lead from atmospheric deposition (sweet corn and tomatoes). Belowground crops are protected from atmospheric deposition but have slightly higher concentrations of lead, partly because lead accumulates in the roots of plants (potatoes, onions, carrots). Leafy aboveground plants (lettuce, spinach, wheat) have even higher lead concentrations presumably because of increased exposure to atmospheric lead. The assumption that can be made here is that, in the absence of atmospheric deposition, exposed aboveground plant parts would have lead concentrations similar to sweet corn and tomatoes.

The data on these ten crops suggest that root vegetables have lead concentrations between 0.0046 and 0.009 μg/g. This is all lead of soil origin, of which presumably half is natural and half anthropogenic (called indirect atmospheric lead here). Aboveground parts not exposed to significant amounts of atmospheric deposition (sweet corn and tomatoes) have less lead internally, also equally divided between natural and indirect atmospheric lead. If it is assumed that this same concentration is the internal concentration for aboveground parts for other plants, it is apparent that five crops (wheat, field corn, rice, spinach, and lettuce) have direct atmospheric deposition in proportion to surface area and estimated duration of exposure. The deposition rate of only 0.04 $ng/cm^2 \cdot day$, which is much smaller than would normally be expected in rural environments (see Section 6.4.1) could account for these amounts of direct atmospheric lead. In this scheme, soybeans are anomalously high. Soybeans grow inside a sheath and should have an internal lead concentration similar to sweet corn.

These discussions lead to the conclusion that root parts and protected aboveground parts of edible crops contain natural lead and indirect atmospheric lead, both derived from the soil. For exposed aboveground parts, any lead in excess of the average found on unexposed aboveground parts is considered to be the result of direct atmospheric deposition.

Near smelters, Merry et al. (1981) found a pattern different from roadside studies cited above. They observed that wheat crops contained lead in proportion to the amount of soil lead, not vegetation surface contamination. A similar effect was reported by Harris (1981).

7.2.2.2.2 Livestock. Lead in forage was found to exceed 950 μg/g within 25 m of roadsides with 15,000 or more vehicles per day (Graham and Kalman, 1974). At lesser traffic densities, 200 μg/g were found. Other reports have observed 20-660 μg/g with the same relationship to traffic density and distance from the road (see review by Graham and Kalman, 1974). A more recent study by Crump and Barlow (1982) showed that the accumulation of lead in forage is directly related to the deposition rate, which varied seasonally according to traffic density. The deposition rate was measured using the moss bag technique, in which bags of moss are exposed and analyzed as relative indicators of deposition flux. Rain was not effective in removing lead from the surface of the moss. The ratio of atmospheric lead to total lead in meat products is partly a function of the same ratio in forage. The fact that most lead in cattle is stored in bones and not eaten by man does not alter the ratio of atmospheric to total lead in meat.

Factors that might add non-atmospheric lead would be soil ingestion by cattle, processed food given to cattle in feedlots, and lead added during processing. Thornton and Abrahams (1983) estimated that 1 to 18 percent of the dry matter ingested by cattle is soil, based the titanium content of feces. Soil ingestion increases when overgrazing is permitted because of dry weather, seasonal changes or other farm management practices. Most of the ingested soil would be from the upper 1-5 cm. In a normal pasture this soil layer would contain a significant fraction of atmospheric lead.

7.2.3 Lead in Surface and Ground Water

Lead occurs in untreated water in either dissolved or particulate form. Dissolved lead is operationally defined as that which passes through a 0.45 µm membrane filter. Because atmospheric lead in rain or snow is retained by soil, there is little correlation between lead in precipitation and lead in streams which drain terrestrial watersheds. Rather, the important factors seem to be the chemistry of the stream (pH and hardness) and the volume of the stream flow. For groundwater, chemistry is also important, as is the geochemical composition of the water-bearing bedrock.

Of the year-round housing units in the United States, 84 percent receive their drinking water from a municipal or private supply of chemically treated surface or ground water. The second largest source is privately owned wells (U.S. Bureau of the Census, 1982). In some communities, the purchase of untreated bottled drinking water is a common practice. The initial concentration of lead in this water depends largely on the source of the untreated water.

7.2.3.1. Typical Concentrations of Lead in Untreated Water

7.2.3.1.1 Surface water. Durum et al. (1971) reported lead concentrations in the range of 1-55 µg/l in 749 surface water samples in the United States. Very few samples were above 50 µg/l, and the average was 3.9 µg/l. Chow (1978) reviewed other reports with mean values between 3 and 4 µg/l. The National Academy of Sciences (1980) reported a mean of 4 µg/l, with a range from below detection to 890 µg/l. Concentrations of 100 µg/l were found near sites of sewage treatment, urban runoff, and industrial waste disposal.

Because 1 µg lead/l was at or below the detection limit of most investigators during the 1970's, it is likely that the mean of 3-4 µg/l was unduly influenced by a large number of erroneously high values at the lower range of detection. On the other hand, Patterson (1980) reports values of 0.006-0.05 µg/l for samples taken from remote streams. Extreme care was taken to avoid contamination and analytical techniques sensitive to less than 0.001 µg/l were used.

Streams and lakes are influenced by their water chemistry and the lead content of their sediments. At neutral pH, lead moves from the dissolved to the particulate form and the particles eventually pass to sediments. At low pH, the reverse pathway generally takes place. Hardness, which is a combination of the Ca and Mg concentration, also can influence lead concentrations. At higher concentrations of Ca and Mg, the solubility of lead decreases. Further discussion of the chemistry of lead in water may be found in Sections 6.5.2.1 and 8.2.2.

7.2.3.1.2 Ground water. Municipal and private wells account for a large percentage of the drinking water supply. This water typically has a neutral pH and somewhat higher hardness than surface water. Lead concentrations are not influenced by acid rain, surface runoff, or atmospheric deposition. Rather, the primary determinant of lead concentration is the geochemical makeup of the bedrock that is the source of the water supply. Ground water typically ranges from 1 to 100 μg lead/l (National Academy of Sciences, 1980). Again, the lower part of the range may be erroneously high due to difficulties of analysis. It is also possible that the careless application of fertilizers or sewage sludge to agricultural lands can cause contamination of ground water supplies.

7.2.3.1.3 Natural vs. anthropogenic lead in water. Although Chow (1978) reports that the natural lead concentration of surface water is 0.5 μg/l, this value may be excessively high. In a discussion of mass balance considerations (National Academy of Sciences, 1980), natural lead was suggested to range from 0.005 to 10 μg/l. Patterson (1980) used further arguments to establish an upper limit of 0.02 μg/l for natural lead in surface water. This upper limit will be used in further discussions of natural lead in drinking water.

Because ground water is free of atmospheric lead, lead in ground water should probably be considered natural in origin as it occurs at the well head, unless there is evidence of surface contamination.

7.2.3.2 Human Consumption of Lead in Water. Whether from surface or ground water supplies, municipal waters undergo extensive chemical treatment prior to release to the distribution system. There is no direct effort to remove lead from the water supply. However, some treatments, such as flocculation and sedimentation, may inadvertently remove lead along with other undesirable substances. On the other hand, chemical treatment to soften water increases the solubility of lead and enhances the possibility that lead will be added to water as it passes through the distribution system.

7.2.3.2.1 Contributions to drinking water. For samples taken at the household tap, lead concentrations are usually higher in the initial volume (first daily flush) than after the tap has been running for some time. Water standing in the pipes for several hours is intermediate between these two concentrations (Sharrett et al., 1982; Worth et al., 1981). Common plumbing materials are galvanized and copper pipe; lead solder is usually used to seal the joints of copper pipes. Lead pipes are seldom in service in the United States, except in the New England states (Worth et al., 1981), and as a flexible fitting between the main line and the house service pipe.

Average lead content of running water at the household tap is generally lower (8 μg/l) than in some untreated water sources (25-30 μg/l) (Sharrett et al., 1982). Water treatment removes lead associated with the suspended solids in raw surface waters. If first flush or standing water is sampled, the lead content may be considerably higher. Sharrett et al. (1982) showed that in both copper and galvanized pipes, lead concentrations were increased by a factor of two when the sample was taken without first flushing the line (see Section 7.3.1.3).

The age of the plumbing is an important factor. New copper pipes with lead solder exposed on the inner surface of the joints produce the highest amount of lead in standing water. After about six years, this lead is either leached away or covered with calcium deposits, and copper pipes subsequently have less lead in standing water than galvanized pipes. Because lead pipes are rarely used in the United States, exposure from this source will be treated as a special case in Section 7.3.2.1.4. The pH of the water is also important; the acid water of some eastern and northwestern United States localities can increase the leaching rate of lead from lead pipes or lead solder joints and prevent the buildup of a protective coating of calcium carbonate plaque.

Table 7-9 summarizes the contribution of atmospheric lead to drinking water. In this determination, the maximum reported value for natural lead (0.02 μg/l) was used, all additional lead in untreated water is considered to be of atmospheric origin, and it is assumed that treatment removes 85 percent of the original lead, and that any lead added during distribution is non-atmospheric anthropogenic lead.

7.2.3.2.2 Contributions to food. The use of treated water in the preparation of food can be a significant source of lead in the human diet. There are many uncertainties in determining this contribution, however. Water used in food processing may be from a municipal supply or a private well. This water may be used to merely wash the food, as with fruits and vegetables, or as an actual ingredient. Water lead may remain on food that is partially or entirely dehydrated during processing (e.g., pasta). Water used for packing or canning may be used with the meal or drained prior to preparation. It is apparent from discussions in Section 7.3.1.3 that, considering both drinking water and food preparation, a significant amount of lead can be consumed by humans from treated water. Only a small fraction of this lead is of atmospheric origin, however.

TABLE 7-9. SUMMARY OF LEAD CONCENTRATIONS IN DRINKING WATER SUPPLIES (μg/l)

Source	Natural lead	Indirect atmospheric lead	Direct atmospheric lead	Non-atmospheric anthropogenic lead	Total lead
Untreated					
Lakes	0.02	15	10	0	25
Rivers	0.02	15	15	0	30
Streams	0.02	2.5	2.5	0	5
Groundwater	3	0	0	0	3
Treated					
Surface	0.003	2.5	1.5	4	8
Ground	0.45	0	0	7.5	8

Source: Text

7.2.4 Summary of Environmental Concentrations of Lead

Lead concentrations in environmental media that are in the pathway to human consumption are summarized in Table 7-10. These values are estimates derived from the preceding discussions. A single value has been used, rather than a range, in order to facilitate further estimates of actual human consumption. This use of a single value is not meant to imply a high degree of certainty in its determination or homogeneity within the human population. The units for water are converted from μg/l as in Table 7-9 to μg/g to facilitate the discussions of dietary consumption of water and beverages.

TABLE 7-10. SUMMARY OF ENVIRONMENTAL CONCENTRATIONS OF LEAD

Medium	Natural lead	Atmospheric lead	Total lead
Urban air (μg/m^3)	0.00005	0.3-1.1	0.3-1.1
Rural air (μg/m^3)	0.00005	0.15-0.3	0.15-0.3
Total soil (μg/g)	8-25	3-5	10-30
Food crops (μg/g)	0.0025	0.00-0.042	0.002-0.045
Surface water (μg/g)*	0.00002	0.005-0.030	0.005-0.030
Ground water (μg/g)*	0.003	0.00	0.001-0.1

*Note change in units from Table 7-9.

Because concentrations of natural lead are generally three to four orders of magnitude lower than anthropogenic lead in ambient rural or urban air, all atmospheric contributions of lead are considered to be of anthropogenic origin. Natural soil lead typically ranges from 10 to 30 µg/g, but much of this is tightly bound within the crystalline matrix of soil minerals at normal soil pHs of 4-8. Lead in the organic fraction of soil is part natural and part atmospheric. The fraction derived from fertilizer is considered to be minimal. In undisturbed rural and remote soils, the ratio of natural to atmospheric lead is about 1:1, perhaps as high as 1:3. This ratio persists in soil moisture and in internal plant tissues. Thus, some of the internal lead in crops is of anthropogenic origin, and some is natural. Information on the effect of fertilizer on this ratio is not available. Lead in untreated surface water is 99 percent anthropogenic. Except near municipal waste outfalls, this anthropogenic lead is mostly atmospheric. It is possible that 75 percent of this lead is removed during treatment. Lead in untreated ground water is presumed to be natural in the absence of evidence of groundwater contamination.

In tracking air lead through pathways to human exposure, it is necessary to distinguish between lead of atmospheric origin that has passed through the soil (indirect atmospheric lead), and atmospheric lead that has deposited directly on crops or water. Because indirect atmospheric lead will remain in the soil for many decades, this source is insensitive to projected changes in atmospheric lead concentrations. Regulation of ambient air lead concentrations will not affect indirect atmospheric lead concentrations over the next several decades.

The method used in this document for calculating the relative contribution of atmospheric lead to total potential human exposure relies partially on the relationship between air concentration and deposition flux described on Section 6.4. Estimates of contributions from other sources are usually based on the observed value for total lead concentration from which the estimated contribution of atmospheric lead is subtracted. The forms of lead subject to the greatest human exposure are atmospheric lead, lead in food cans, and lead in paint pigments. There is little evidence for the substantial contribution of other forms of anthropogenic lead to the total lead consumption by the general U.S. population.

7.3 POTENTIAL PATHWAYS TO HUMAN EXPOSURE

The preceding section discussed ambient concentrations of lead in the environment, focusing on levels in the air, soil, food crops, and water. In this section, environmental lead concentrations are examined from the perspective of pathways to human exposure (Figure 7-1). Exposure is a measure of the amount of pollutant available at the interface between the human and the human environment. The estimation of exposure requires a knowledge of pollutant concentrations of each environmental component, the amounts of each environmental component consumed, and the time budgets or other specific activities normal for humans (Moschandreas, 1981). For this analysis, a current baseline exposure scenario is described for an individual with a minimum amount of daily lead consumption. It is assumed that this person lives and works in a nonurban environment, eats a normal diet of food taken from a typical grocery shelf, and has no habits or activities that tend to increase lead exposure. Without drastic changes in lifestyle, lead exposure at the baseline level is considered unavoidable without further reductions of lead in the atmosphere or in canned foods. Most of the baseline lead is of anthropogenic origin, although a portion is natural, as discussed in Section 7.3.1.5.

7.3.1 Baseline Human Exposure

To arrive at a minimum or baseline exposure for humans, it is necessary to begin with the environmental components (air, soil, food crops, and water) that are the major sources of lead consumed by humans (Table 7-10). These components are measured frequently, even monitored routinely in the case of air, so that many data are available on their concentrations. But there are several factors that modify these components prior to actual human exposure. We do not breathe air as monitored at an atmospheric sampling station. We may be closer to or farther from the source of lead than is the monitor. We may be inside a building, with or without filtered air; the water we drink does not come directly from a stream or river. It has passed through a chemical treatment plant and a distribution system. A similar type of processing has modified the lead levels present in our food.

It is inappropriate to assess human exposure to lead from a single source or through a single medium without a simultaneous assessment from other sources (Laxon et al., 1985). Our ability to monitor the environment depends on the available technology. But our knowledge of human exposure depends on the correct understanding of the transfer of a pollutant from the environmental component to the human body. In the past, exposure to air pollutants have been interpreted strictly in the context of inhalation, with little consideration given to other routes of exposure. This document attempts to assess the total human exposure to lead from all sources and through all pathways.

Besides the atmospheric lead in environmental components, there are two other anthropogenic sources that contribute to this baseline of human exposure: paint pigments and lead solder (Figure 7-7). Solder contributes directly to the human diet through canned food and copper water distribution systems. Chips of paint pigments are discussed later under special environments. But paint and solder are also a source of lead-bearing dusts. The most common dusts in the baseline human environment are street dusts and household dusts. They originate as emissions from mobile or stationary sources, as the oxidation products of surface exposure,

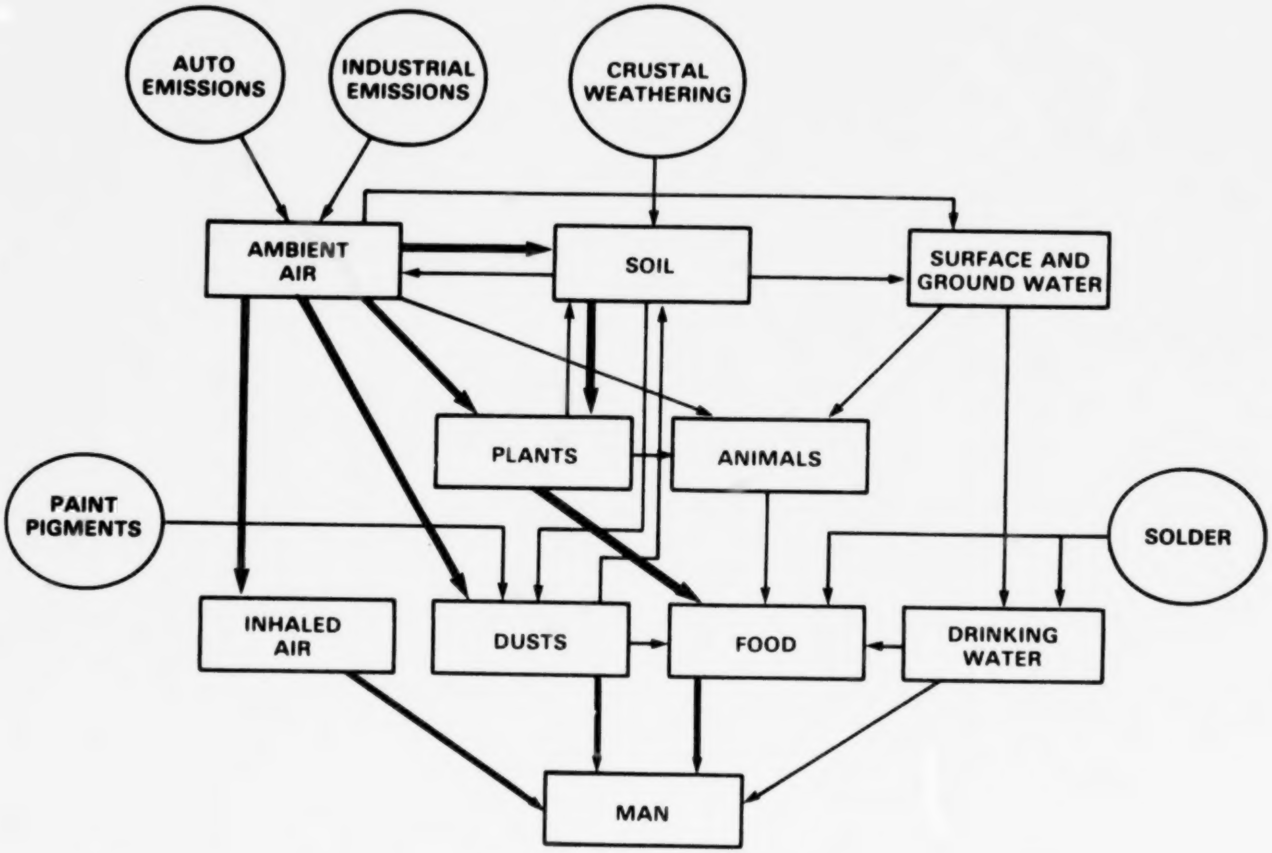
AUTO EMISSIONS
INDUSTRIAL EMISSIONS
CRUSTAL WEATHERING
AMBIENT AIR
SOIL
SURFACE AND GROUND WATER
PLANTS
ANIMALS
PAINT PIGMENTS
SOLDER
INHALED AIR
DUSTS
FOOD
DRINKING WATER
MAN

Figure 7-7. Paint pigments and solder are two additional sources of potential lead exposure which are not of atmospheric origin. Solder is common even in baseline exposures and may represent 30 to 45 percent of the baseline human consumption. Paint pigments are encountered in older houses and in soils adjacent to older houses.

or as products of frictional grinding processes. Dusts are different from soil in that soil derives from crustal rock and typically has a lead concentration of 10-30 µg/g, whereas dusts come from both natural and anthropogenic sources and vary from 1,000 to 10,000 µg/g.

The discussion of the baseline human exposure traces the sequence from ambient air to inhaled air, from soil to prepared food, from natural water to drinking water, and from paint, solder and aerosol particles to dusts. At the end of this section, Table 7-18 summarizes the four sources by natural and anthropogenic contributions, with the atmospheric contribution to the anthropogenic fraction identified. Reference to this table will guide the discussion of human exposure in a logical sequence that ultimately presents an estimate of the exposure of the human population to atmospheric lead. To construct this table, it was necessary to make decisions based on sound scientific judgment, extracted from the best available data. This method provides a working approach to identifying sources of lead that can be easily modified as more accurate data become available.

7.3.1.1 Lead in Inhaled Air. A principal determinant of atmospheric lead concentration is distance from the source. At more than 100 m from a major highway or more than 2 km from a stationary source, lead concentrations generally drop to constant levels (see Section 6.3), and the particle size distribution shifts from a bimodal distribution to a unimodal one with an MMAD of about 0.2 µm. Because the concentration of atmospheric lead at nonurban stations is generally 0.05-0.15 µg/m^3, a value of 0.1 µg/m^3 may reasonably be assumed. A correction can be made for the indoor/outdoor ratio assuming the average individual spends 20-22 hours/day in an unfiltered inside atmosphere and the average indoor/outdoor ratio for a nonurban location is 0.5 (Table 7-6). The adjusted air concentration becomes 0.05 µg/m^3 for baseline purposes.

The concentration of natural lead in the atmosphere, discussed in Section 7.2.1.1.3, is probably about 0.00005 µg/m^3. This is an insignificant amount compared to the anthropogenic contribution of 0.2 µg/m^3. A summary of lead in inhaled air appears in Table 7-11.

TABLE 7-11. SUMMARY OF INHALED AIR LEAD EXPOSURE

Population	Adjusted air Pb conc.* (µg/m^3)	Amount inhaled (m^3/day)	Total lead exposure (µg/day)	Natural Pb (µg/day)	Direct atmospheric Pb (µg/day)
Children (2 year-old)	0.05	10	0.5	0.001	0.5
Adult, working inside	0.05	20	1.0	0.002	1.0
Adult, working outside	0.10	20	2.0	0.004	2.0

*Values adjusted for indoor/outdoor ratio of lead concentrations and for daily time spent outdoors.

Source: Text

7.3.1.2 Lead in Food. The route by which many people receive the largest portion of their daily lead intake is through foods. Several studies have reported average dietary lead intakes in the range 100 to 500 µg/day for adults, with individual diets covering a much greater range (Schroeder and Tipton, 1968; Mahaffey, 1978; Nutrition Foundation, Inc. 1982). Gross (1981) analyzed results of the extensive lead mass balance experiments described by Kehoe (1961), which were conducted from 1937 to 1972. According to these data, total dietary lead intake decreased from approximately 300 µg/day in 1937 to 100 µg/day in 1970, although there is considerable variability in the data. Only a fraction of this lead is absorbed, as discussed in Chapter 10.

The amount of lead typically found in plants and animals is discussed in Section 7.2.2.2. The sources of this lead are air, soil, and untreated waters (Figure 7-1). Food crops and livestock contain lead in varying proportions from the atmosphere and natural sources. From the farm to the dinner table, lead is added to food as it is harvested, transported, processed, packaged, and prepared. The sources of this lead are dusts of atmospheric and industrial origin, metals used in grinding, crushing, and sieving, solder used in packaging, and water used in cooking.

The American diet is extremely complex and variable among individuals. Pennington (1983) has described the basic diets, suppressing individual variation but identifying 234 typical food categories, for Americans grouped into eight age/sex groups. These basic diets are the foundation for the Food and Drug Administration's revised Total Diet Study, often called the "market basket" study, beginning in April, 1982. The diets used for this document include food, beverages, and drinking water for 2-year-old children, teen-age males and females, adult males and females (25-30 years of age), and adult males and females (60 - 65 years of age). The 201 typical food categories that constitute the basic diets are an aggregation of 3500 categories of food actually consumed by participants in the two surveys that formed the basis of the Pennington study. Lead concentration data are given for each of these 201 food categories in Table 7D-1 of Appendix 7D and are from a preliminary report of the 1982 and 1983 Total Diet Study provided by the U.S. Food and Drug Administration for the purpose of this document.

In 1982, the Nutrition Foundation published an exhaustive study of lead in foods, using some data from the National Food Processors Assocation and some data from Canadian studies by Kirkpatrick et al. (1980) and Kirkpatrick and Coffin (1974, 1977). A summary of the available data for 1973-1980 was prepared in an internal report to the FDA prepared by Beloian and McDowell (1981). Portions of these reports were used to interpret the contributions of lead to food during processing.

The following section evaluates the amounts of lead added during each step of the process from the field to the dinner table. In the best case, reliable data exist for the specific situation in question and conclusions are drawn. In some cases, comparable data can be used with a few reasonable assumptions to formulate acceptable estimates of lead contributions. For a portion of the diet, there are no acceptable data and the contributions of lead must, for the time, be listed as of undetermined origin.

7.3.1.2.1 Lead added during handling and transportation to processor. Between the field and the food processor, lead is added to the food crops. It is assumed that this lead is all of direct atmospheric origin. Direct atmospheric lead can be lead deposited directly on food materials by dry deposition, or it can be lead on dust that has collected on other surfaces, then transferred to foods. For the purposes of this discussion, it is not necessary to distinguish between these two forms, as both are a function of air lead concentration.

There are no clear data on how much lead is added during transportation, but some observations are worth noting. First, some fresh vegetables (e.g., potatoes, lettuce, carrots, onions) undergo no further processing other than trimming, washing and packaging. If washed, water without soap is used; no additives or preservatives are used. An estimate of the amount of atmospheric lead added during handling and transportation of all food crops can be made from the observed increases in lead on those fresh vegetables where handling and transportation would be the only source of added lead. Because atmospheric lead deposition is a function of time, air concentration, and exposed surface area, there is an upper limit to the maximum amount of direct atmospheric lead that can be added, except by the accumulation of atmospheric dusts.

7.3.1.2.2 Lead added during preparation for packaging. For some of the canned food items, data are available on lead concentrations just prior to the filling of cans. In the case where the food product has not undergone extensive modification (e.g., cooking, added ingredients), the added lead was most likely derived from the atmosphere or from the machinery used to handle the product. As with transportation, the addition of atmospheric lead is limited to reasonable amounts that can be added during exposure to air, and reasonable amounts of atmospheric dust accumulation on food processing surfaces. One process that may increase the exposure of the food to air is the use of air in separating food items, as in wheat grains from chaff.

Where modification of the food product has occurred, the most common ingredients added are sugar, salt, and water. It is reasonable that water has a lead concentration similar to drinking water reported in Section 7.3.1.3 (0.008 μg/g) and that sugar (Boyer and Johnson, 1982) and salt have lead concentrations of 0.01 μg/g. Grinding, crushing, chopping, and cooking may add lead from the metallic parts of machinery and from industrial greases. A summary of the data (Table 7-12) indicates that about 30 percent of the total lead in canned goods is the result of prepacking processes.

TABLE 7-12. ADDITION OF LEAD TO FOOD PRODUCTS*
(μg/g fresh weight)

Food	In the field (A)	After preparation for packaging (B)	After packaging (C)	After kitchen preparation (D)	Total lead added after harvest (E)
Soft Packaged					
Wheat	0.037	N/A	0.065	--	--
Field corn	0.022	N/A	0.14	0.025	0.003
Potatoes	0.009	N/A	0.018	0.02	0.011
Lettuce	0.013	N/A	0.07	0.015	0.002
Rice	0.007	N/A	0.10	0.084	0.077
Carrots	0.009	N/A	0.05	0.017	0.008
Beef	0.01	N/A	0.07	0.035	0.025
Pork	0.06	N/A	0.10	0.06	--
Metal cans					
Sweet corn	0.003	0.04	0.27	0.28	0.28
Tomatoes	0.002	0.06	0.29	--	--
Spinach	0.045	0.43	0.68	0.86	0.82
Peas	N/A	0.08	0.19	0.22	0.14
Applesauce	N/A	0.08	0.24	0.17	0.09
Apricots	N/A	0.07	0.17	--	0.10
Mixed fruit	N/A	0.08	0.24	0.20	0.12
Plums	N/A	0.09	0.16	--	0.07
Green beans	N/A	0.16	0.32	0.16	--

*This table summarizes the stepwise addition of lead to food products at several stages between the field and the dinner table. Data in column A are from Wolnik et al. (1983, 1985), columns B and C from National Food Processors Association (1982), and column D from U.S. FDA (1985). Column E is calculated as column D - column A. Where data are not available in column A, the values in column B were used. For the most part, column C values closely approximate column D values, even though they are from separate studies, suggesting most of the lead in food production is added prior to kitchen preparation.

N/A: data not available.

Occasionally, the processing or preparation of food may separate lead into a single product or byproduct. Hayashi et al. (1982) found that lead in milk is isolated during processing from butter and ends up in the buttermilk. Thus lead in butter is typically lower than and buttermilk higher than normal sweet milk. Smart et al. (1981) have found that foods cooked in water adsorb the lead in that water. Consequently, when pasta or similar items are cooked then drained, the lead content of the prepared food is the sum of the dry food and the water. Conversely, when only the water is retained, as with tea bags, the final beverage may have less lead than the original water.

7.3.1.2.3 Lead added during packaging. From the time a product is packaged in bottles, cans or plastic containers, until it is opened in the kitchen, it may be assumed it receives atmospheric lead. Most of the lead that is added during this stage comes from the solder used to seal some types of cans. Estimates by the U.S. FDA, prepared in cooperation with the National Food Processors Association, suggest that lead in solder contributes more than 66 percent of the lead in canned foods where a lead solder side seam is used. This lead was thought to represent a contribution of 20 percent to the total lead consumption in foods (F.R., 1979 August 31).

The full extent of the contribution of the canning process to overall lead levels in albacore tuna was reported in a benchmark study by Settle and Patterson (1980). Using rigorous clean laboratory procedures, these investigators analysed lead in fresh tuna, as well as in tuna packaged in soldered and unsoldered cans. The data, presented in Table 7-13, show that lead concentrations in canned tuna are elevated above levels in fresh tuna by a factor of 4,000, and by a factor of 40,000 above natural levels of lead in tuna. Nearly all of the increase results from leaching of the lead from the soldered seam of the can; tuna from an unsoldered can is elevated by a factor of only 20 compared with tuna fresh from the sea. Note that when fresh tuna is dried and pulverized, as in the National Bureau of Standards reference material, lead levels are seen to increase by a factor of 400 over fresh sea tuna. Table 7-13 also shows the results of analyses conducted by the National Marine Fisheries Service.

TABLE 7-13. PREHISTORIC AND MODERN CONCENTRATIONS IN HUMAN FOOD FROM A MARINE FOOD CHAIN (ng/g fresh weight)

Source	Estimated prehistoric	Modern
Surface seawater	0.0005	0.005
Albacore muscle, fresh	0.03	0.3
Albacore muscle from die-punched unsoldered can	--	7.0
Albacore muscle, lead-soldered can	--	1400
Anchovy from albacore stomach	2.1	21
Anchovy from lead-soldered can	--	4200

Source: Settle and Patterson (1980).

7.3.1.2.4 *Lead added during kitchen usage and storage*. Although there have been several studies of the lead concentrations in food after typical meal preparation, most of the data are not amenable to this analysis because there are no data on lead concentrations before meal preparation. As a part of its compliance program, the U.S. FDA has conducted the Total Diet Study of lead and other trace contaminants in kitchen-prepared food each year since 1973. Because the kitchen-prepared items were composited by category, there is no direct link between a specific food crop and the dinner table. Since April, 1982, this survey has analyzed each food item individually (Pennington, 1983).

Other studies that reflect contributions of lead added during kitchen preparation have been conducted. Capar (1978) showed that lead in acidic foods that are stored refrigerated in open cans can increase by a factor of 2-8 in five days if the cans have a lead-soldered side seam not protected by an interior lacquer coating. Comparable products in cans with the lacquer coating or in glass jars showed little or no increase.

7.3.1.2.5 *Recent changes in lead in food*. As a part of its program to reduce the total lead intake by children (0-5 years of age) to less than 100 µg/day by 1988, the U.S. FDA estimated lead intakes for individual children in a large-scale food consumption survey (Beloian and McDowell, 1981). To convert the survey of total food intakes into lead intake, 23 separate government and industry studies, covering 1973-78, were statistically analyzed. In spite of the variability that can occur among individuals grouped by age, the authors estimated a baseline (1973-78) daily lead intake of 15 µg/day for infants aged 0-5 months, 59 µg/day for children 6-23 months, and 82 µg/day for children 2-5 years. Between 1973 and 1978, intensive efforts were made by the food industry to remove sources of lead from infant food items. By 1980, there had been a 47 percent reduction in the lead concentration for food consumed by the age group 0-5 months and a 7 percent reduction for the 6-23 month age group (Table 7-14). Most of this reduction was accomplished by the discontinuation of soldered cans used for infant formula.

The 47 percent reduction in dietary lead achieved for infants prior to 1980 came about largely because there are relatively few manufacturers of foods for infants and it was comparatively simple for this industry to mount a coordinated program in cooperation with the FDA. There has not yet been a similar decrease in adult foods (Table 7-14) because only a few manufacturers have switched to lead-free cans. As the switchover increases, lead in canned food should decrease to a level as low as 30 percent of the pre-1978 values, and there should be a corresponding decrease of lead in the total adult diet, perhaps as much as 20 percent. The use of lead-soldered cans in the canning industry has decreased from 90 percent in 1979 to 63 percent in 1982. Within the next few years, the two leading can manufacturers expect to produce no more lead-soldered cans for the food industry. A two-year time lag is expected before

TABLE 7-14. RECENT TRENDS OF MEAN LEAD CONCENTRATIONS IN CANNED ADULT AND INFANT FOOD ITEMS (μg/g)

	Early 70's	1976-77	1980-81	1982
Canned food*				
Green beans	0.32	N/A	0.32	0.16
Beans w/pork	0.64	N/A	0.26	0.17
Peas	0.43	N/A	0.19	0.22
Tomatoes	0.71	N/A	0.29	0.21
Beets	0.38	N/A	0.24	0.12
Tomato juice	0.34	N/A	0.08	0.067
Applesauce	0.32	N/A	0.04	0.17
Citrus juice	0.14	N/A	0.11	0.04
Infant food				
Formula concentrate	0.10	0.055	0.01	N/A
Juices	0.30	0.045	0.015	N/A
Pureed foods	0.15	0.05	0.02	N/A
Evaporated milk	0.52	0.10	0.07	N/A

*Boyer and Johnson (1982); 1982 data from U.S. Food and Drug Administration 1985 (see Appendix 7D).

†Data from early 70's and 1976-79 from Jelinek (1982); 1980-81 data from Schaffner (1981).

N/A = data not available.

the last of these cans disappears from the grocery shelf. Some of the 23 smaller manufacturers of cans have announced similar plans over a longer period of time. It is likely that any expected decrease in the contribution of air lead to foods will be complemented by a decrease in lead from soldered cans.

7.3.1.2.6 Summary of lead in food. There are two major sources of lead in food and beverages: atmospheric lead and lead from cans with lead soldered seams. The data of Wolnik et al. (1983, 1985) provide some insight into the amount of atmospheric lead on food crops (Table 7-8). The FDA analyses of foods by category (Table 7D-1 in appendix 7D) clearly show the influence of solder on canned foods compared to fresh foods of the same type. The total food consumption data of Pennington (1983) for 201 adult food categories were multiplied by the mean lead concentrations from Table 7D-1 to determine the total daily exposure of seven age/sex categories of Americans to lead in food and beverages.

For each food category, a separate source coefficient was assigned for direct atmospheric, solder and metallic, indirect atmospheric, and natural lead. Any fraction of lead that could not be otherwise assigned was considered lead of undetermined origin. In this manner, the lead content of 201 food categories was determined for five sources. To simplify the presentation of this data, the 201 food categories have been combined into nine groups based on the scheme of Table 7D-2 in appendix 7D-2 in appendix 7D. The nine categories were specifically selected to emphasize the most probable source of lead. Therefore canned foods were placed in one category to isolate metallic lead, and crop foods in another to isolate atmospheric lead.

The total consumption for the seven age/sex categories and nine food categories is shown in Table 7-15, adapted from Pennington (1983). The amount of lead that is consumed with the food and beverages in Table 7-15 is shown on Table 7-16. This calculation is based solely on the average lead concentrations of each food item of Table 7D-1, the data provided by FDA. To determine the source of this lead, the individual source coefficients for each food item were multiplied by the average lead concentration in Table 7D-1 and by the amount consumed (Pennington, 1983) to get the amount of lead consumed from each source for each age/sex category. An average was taken of each age/sex category then the 201 food items were condensed into the nine food categories and presented on Table 7-17.

TABLE 7-15. TOTAL CONSUMPTION, BY AGE AND SEX, OF FOOD AND BEVERAGES (g/day)

Major food category	Child* 2 yrs	Female	Male	Female	Male	Female	Male
		14-16 yrs		25-30 yrs		60-65 yrs	
Dairy products	390	405	645	245	351	208	279
Meat products	133	182	269	194	319	172	252
Food crops	282	386	528	390	518	437	532
Canned food	72	77	104	73	103	99	119
Canned juices	54	28	30	28	27	17	12
Frozen juices	65	53	75	66	73	72	61
Soda	65	232	274	228	315	78	85
Canned beer	0	0	17	51	318	18	116
Water†	441	596	743	903	1061	1166	1244
Totals	1502	1959	2685	2178	3086	2267	2700

*7.5 g baby food and infant formula were not included in this evaluation.

†Includes coffee, tea, and powdered drinks.

Source: Data are summarized from Pennington (1983) according to Table 7D-2.

TABLE 7-16. TOTAL CONSUMPTION, BY AGE AND SEX, OF LEAD IN MILK AND FOOD AND BEVERAGES (μg/day)

Major food category	Child* 2 yrs	Female 14-16 yrs	Male 14-16 yrs	Female 25-30 yrs	Male 25-30 yrs	Female 60-65 yrs	Male 60-65 yrs
Dairy products	2.8	3.5	5.4	2.5	3.4	2.3	3.1
Meat products	3.4	4.8	7.4	5.0	7.4	4.0	5.4
Food crops	5.5	8.1	11.7	7.9	11.3	7.8	9.6
Canned food	7.3	8.1	11.8	8.8	12.0	11.6	14.4
Canned juices	2.7	1.4	1.5	1.4	1.4	0.9	0.6
Frozen juices	0.5	0.5	0.7	0.6	0.7	0.7	0.5
Soda	0.7	2.3	3.0	2.1	2.9	0.9	0.9
Canned beer	0.0	0.0	0.1	0.7	2.5	0.3	1.0
Water†	2.1	2.5	3.2	3.0	3.6	3.9	4.2
Totals	25.0	31.2	44.8	32.0	45.2	32.4	39.7

†Includes coffee, tea, and powdered drinks.

It is apparent that about 43 percent of lead in food and beverages milk and food can be attributed to direct atmospheric deposition, compared to 42 percent from solder or other metal sources. Of the remaining 5 percent for which the source is as yet undetermined, it is likely that further research will show this lead to be part atmospheric in origin and part from solder and other industrial metals.

This dietary lead consumption is used to calculate the total baseline human exposure in Section 7.3.1.5 and is the largest baseline source of lead. Possible additions to dietary lead consumption are discussed in Section 7.3.2.1.3 with respect to urban gardens.

Because the U.S. FDA is actively pursuing programs to decrease lead in adult foods, it is probable that there will be a decrease in total dietary lead consumption over the next decade independent of projected decreases in atmospheric lead concentration. With both sources of lead minimized, the lowest reasonable estimated dietary lead consumption would be 10-15 μg/day for adults and children. This estimate is based on the assumption that about 90 percent of the direct atmospheric lead, solder lead, and lead of undetermined origin would be removed from the diet, leaving 8 μg/day from these sources and 3 μg/day of natural and indirect atmospheric lead.

TABLE 7-17. SUMMARY BY SOURCE OF LEAD CONSUMED FROM FOOD AND BEVERAGES (μg/day)

Major food category	Total lead*	Natural lead	Atmospheric lead		Lead from solder and other metals	Lead of undetermined origin
			indirect	direct		
Dairy	3.3	0.030	0.030	2.74	0.00	0.50
Meat	5.3	0.040	0.040	4.11	0.41	0.70
Food crops	8.8	0.880	0.880	6.60	0.00	0.44
Canned foods	10.6	0.120	0.120	0.92	9.40	0.04
Canned juices	1.4	0.001	0.062	0.04	1.30	0.00
Frozen Juices	0.6	0.001	0.110	0.07	0.42	0.00
Soda	1.8	0.005	0.280	0.21	1.30	0.00
Canned Beer	0.7	0.001	0.140	0.05	0.51	0.00
Water	3.2	0.010	0.850	0.54	1.80	0.00
Total	35.7	1.088	2.512	15.28	15.14	1.68
Percent		3.0%	7.0%	42.8%	42.4%	4.7%

*Based on average lead consumption by 7 age/sex groups. There may be some direct atmospheric lead and solder lead in the category of undetermined origin.

7.3.1.3 Lead in Drinking Water. The U.S. Public Health Service standards specify that lead levels in drinking water should not exceed 50 μg/l. The presence of detectable amounts of lead in untreated public water supplies was shown by Durum et al. (1971) to be widespread, but only a few samples contained amounts above the 50 μg/l standard.

The major source of lead contamination in drinking water is the water distribution system. Water that is corrosive can leach considerable amounts of lead from lead plumbing and lead compounds used to join pipes. Moore (1977) demonstrated the effect of water standing in pipes overnight. Lead concentrations dropped significantly with flushing at 10 l/min for five minutes (Figure 7-8). Lead pipe currently is in use in some parts of New England for water service lines and interior plumbing, particularly in older urban areas. The contributions of lead plumbing to potential human exposure are considered additive rather than baseline and are discussed in Section 7.3.2.1.4.

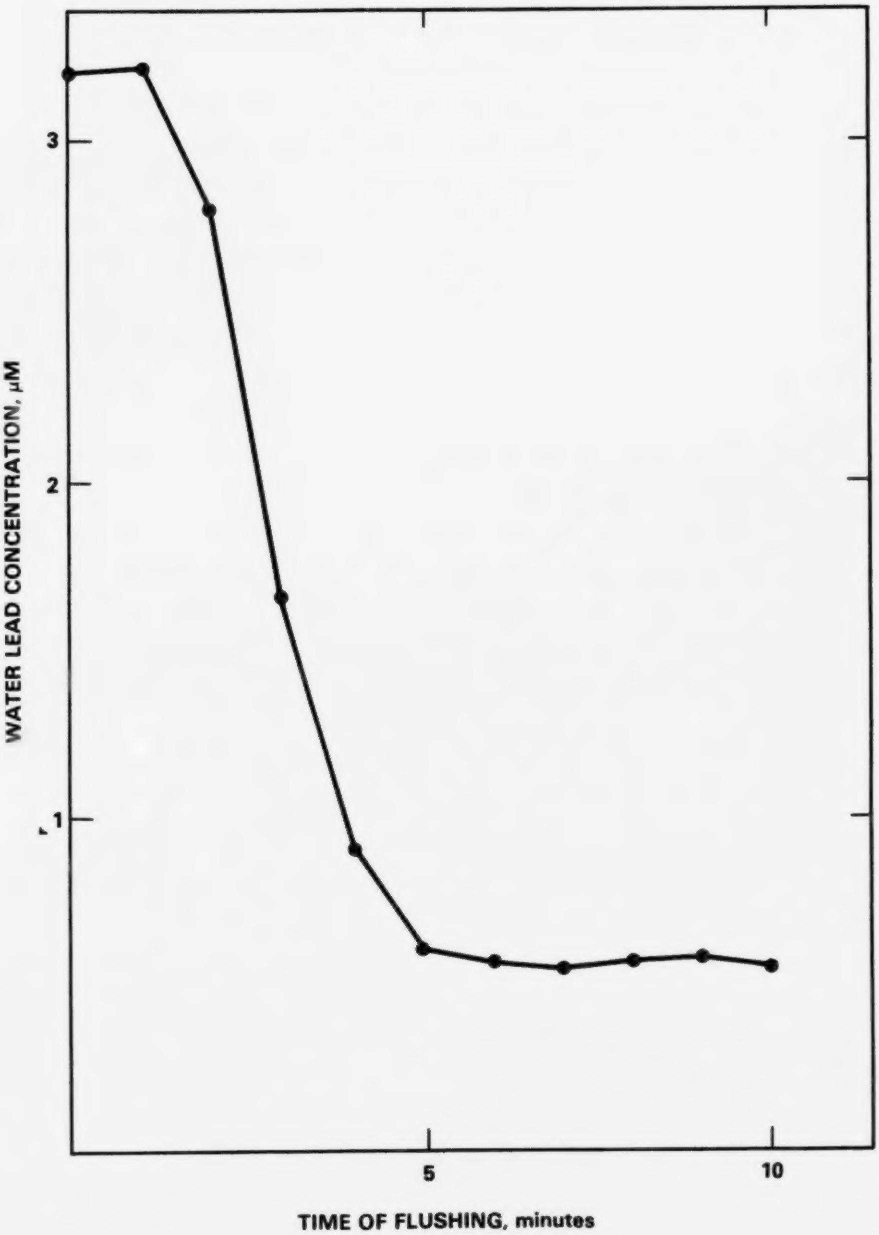

Figure 7-8. Change in drinking water lead concentration in a house with lead plumbing for the first use of water in the morning. Flushing rate was 10 liters/minute.

Source: Moore (1977).

There have been several studies in North America and Europe of the sources of lead in drinking water. A recent study in Seattle, WA by Sharrett et al. (1982) showed that the age of the house and the type of plumbing determined the lead concentration in tap water. Standing water in copper pipes from houses newer than five years averaged 31 µg/l; those less than 18 months average about 70 µg/l. Houses older than five years and houses with galvanized pipe averaged less than 6 µg/l. The source of the water supply, the length of the pipe, and the use of plastic pipes in the service line had little or no effect on the lead concentrations. It appears certain that the source of lead in new homes with copper pipes is the solder used to join these pipes, and that this lead is either leached away with age or isolated by accumulated deposits within the pipes. A study of copper pipes in cottages using local lakes for a water source revealed a similar pattern of lead with increased standing time in the pipes (Meranger et al., 1983). The lead concentration in the first liter drawn continued to increase with standing time, even up to ten days.

The Sharrett et al. (1982) study of the Seattle population also provided data on water and beverage consumption which extended the scope of the Pennington (1983) study of all Americans. While the total amount of liquids consumed was slightly higher in Seattle (2200 g/day vs. 1800 g/day for all Americans), the breakdown between water consumed inside and outside the home can prove useful. Men, women, and children consume 53, 87, and 87 percent, respectively, of their water and beverages within the home.

Bailey and Russell (1981) have developed a model for population exposure to lead in home drinking water. The model incorporates data for lead concentration as a function of stagnation time in the pipes, as well as probability distributions for times of water use throughout the day. Population surveys conducted as part of the United Kingdom Regional Heart Survey provided these water-use distributions.

Other studies have been conducted in Canada and Belgium. Lead levels in water boiled in electric kettles were measured in 574 households in Ottawa (Wigle and Charlebois, 1978). Concentrations greater than 50 µg/l were observed in 42.5 percent of the households, and excessive lead levels were associated with kettles more than five years old.

7.3.1.4 Lead in Dusts. By technical definition, dusts are solid particles produced by the disintegration of materials (Friedlander, 1977) and appear to have no size limitations. Although dusts are of complex origin, they may be placed conveniently into a few categories relating to human exposure. Generally, the most convenient categories are household dusts, soil dust, street dusts, and occupational dusts. In each case, the lead in dust arises from a complex mixture of fine particles of soil, flaked paint, and airborne particles of industrial or automotive origin. It is a characteristic of dust particles that they accumulate on exposed surfaces and are trapped in the fibers of clothing and carpets. Ingestion of dust particles, rather than inhalation, appears to be the greater problem in the baseline environment, especially ingestion during meals and playtime activity by small children.

Two other features of dust are important. First, they must be described in both concentration and amount. The concentration of lead in street dust may be the same in a rural and urban environment, but the amount of dust may differ by a wide margin. Secondly, each category represents a different combination of sources. Household dusts contain some atmospheric lead, some paint lead, and some soil lead. Street dusts contain atmospheric, soil, and occasionally paint lead. This apparent paradox does not prevent the evaluation of exposures to dust, but it does confound efforts to identify the amounts of atmospheric lead contributed to dusts. For the baseline human exposure, it is assumed that workers are not exposed to occupational dusts, nor do they live in houses with interior leaded paints. Street dust, soil dust, and some household dust are the primary dust sources for baseline potential human lead exposure.

In considering the impact of street dust on the human environment, the obvious question arises as to whether lead in street dust varies with traffic density. In a transect through Minneapolis/St. Paul, Mielke et al. (1984) found soil lead concentrations 10 to 1000-fold higher near major interstate highways. Nriagu (1978) reviewed several studies of lead in street dust. Warren et al. (1971) reported 20,000 µg Pb/g street dust in a heavily trafficked area. In the review by Nriagu (1978), street dust lead concentrations ranged from 300 to 18,000 µg/g in several cities in the United States. More recent studies have attempted to characterize lead in street dust in greater detail. Franz and Hadley (1981) separated street dust by particle size and found that smaller particles contain greater concentrations of lead. One-third of the mass was less than 150 µm and contained 37 percent of the total lead. The average concentration in the Albuquerque street dust was 5000 µg/g, 20 percent of which was attributed to curb paint. Dong et al. (1984) separated street dusts by mechanical sieve and found, with one exception, 50 percent or more of the lead on clay-sized particles, the smallest fraction both in particle size and in total mass (5-6 percent). There was, however, considerable variation in the absolute concentrations of lead in the samples from the same location taken four days apart.

There are several reports of street dusts outside the United States that show similar relationships. Fergusson and Ryan (1984) found concentrations in small urban cities in Canada, New Zealand and Jamaica ranged from 700 to 2000 µg/g, while in New York and London the range was from 2000 to 4000 µg/g. Sequential extractions showed much of the lead (44 percent) was on the Fe-Mn oxide fraction, but that 36 percent was on the exchangeable and carbonate fractions that are more readily available. Gibson and Farmer (1984) also found 41 percent of the street dust lead in Glascow, Scotland to be on the exchangeable and carbonate fractions.

Duggan (1984) attempted to relate London street dust lead to airborne lead concentrations and found that airborne variations with time were greater than for dust, but spatial variations were greater for dust. The results suggested that dust may be an adequate measure of long-term (three month) ambient concentrations, but that several samples over a wide area must be taken. In a related study, Thornton et al. (1985) recommended the adoption of guidelines for urban dust lead concentrations to the Greater London Council. The recommendation was that lead concentrations of 500 µg/g in the fraction smaller than 0.5 mm justified further investigation, whereas concentrations above 5000 µg/g justified control measures. Duggan et al. (1985) reported that the amount of lead on children's hands was proportional to the concentration of lead in playground dust. This relationship was nearly linear up to 4000 µg Pb/g dust.

In Hong Kong, lead in street dust ranged from 960 to 7400 µg/g with no direct relationship to traffic volume (Ho, 1979). In other reports from Hong Kong, Lau and Wong (1982) found values from 130 µg/g at 20 vehicles/day to 3900 µg/g at 37,000 vehicles/day. Fourteen sites in this study showed close correlation with traffic density.

In the United Kingdom, lead in urban and rural street dusts was determined to be 970 and 85 µg/g, respectively, by Day et al. (1975). A later report by this group (Day et al., 1979) discusses the persistency of lead dusts in rainwashed areas of the United Kingdom and New Zealand and the potential health hazard due to ingestion by children. They concluded that, whereas the acidity of rain was insufficient to dissolve and transport lead particles, the potential health hazard lies with the ingestion of these particles during the normal play activities of children residing near these areas. A child playing at a playground near a roadside might consume 20-200 µg lead while eating a single piece of candy with unwashed hands. It appears that in nonurban environments, lead in street dust ranges from 80 to 130 µg/g, whereas urban street dusts range from 1,000 to 20,000 µg/g. For the purpose of estimating potential human exposure, an average lead value of 90 µg/g in street dust is assumed for baseline exposure on Table 7-18, and 1500 µg/g in the discussions of urban environments in Section 7.3.2.1.

Dust is also a normal component of the home environment. It accumulates on all exposed surfaces, especially furniture, rugs and windowsills. For reasons of hygiene and respiratory health, many homemakers take great care to remove this dust from the household. Because there are at least two circumstances where these measures are inadequate, it is important to consider the possible concentration of lead in these dusts in order to determine potential exposure to young children. First, some households do not practice regular dust removal, and secondly, in some households of workers exposed occupationally to lead dusts, the worker may carry dust home in amounts too small for efficient removal but containing lead concentrations much higher than normal baseline values.

TABLE 7-18. CURRENT BASELINE ESTIMATES OF POTENTIAL HUMAN EXPOSURE TO DUSTS

	Dust lead conc. (µg/g)	Dust ingested (g/day)	Dust lead consumed (µg/day)	Source of lead		
				Natural (µg/day)	Atmos. (µg/day)	Undetermined (µg/day)
Child						
Household dusts	300	0.05	15	0.5	14.5	0.0
Street dust	90	0.04	4.5	0.0	4.5	0.0
Occupational dust	150	0.01	1.5	0.1	0.0	1.4
Total		0.10	21.0	0.6	19.0	1.4
Percent			100%	2.8%	90.5%	6.7%
Adult						
Household dusts	300	0.01	3	0.1	2.9	0.0
Street dust	90	0.0	0	0.0	0.0	0.0
Occupational dust	150	0.01	1.5	0.1	0.0	1.4
Total		0.02	4.5	0.2	2.9	1.4
Percent			100%	4.5%	64.4%	31.1%

In Omaha, Nebraska, Angle and McIntire (1979) found that lead in household dust ranged from 18 to 5600 µg/g. Clark et al. (1985) found household dusts in Cincinnati ranged from 70 to 16000 µg/g, but that much of the variations could be attributed to housing quality. Public housing averaged 350 µg/g, rehabilitated 600 µg/g, and averages in private housing ranged from 1400 to 3000 based on external estimates of condition from satisfactory to deteriorating to dilapidated. In Lancaster, England, a region of low industrial lead emissions, Harrison (1979) found that household dust ranged from 510 to 970 µg/g, with a mean of 720 µg/g. They observed that dust contained soil particles (10-200 µm in diameter), carpet and clothing fibers, animal and human hairs, food particles, and an occasional chip of paint. The previous Lead Criteria Document (U.S. Environmental Protection Agency, 1977) summarized earlier reports of lead in household dust showing residential suburban areas ranging from 280 to 1,500 µg/g, urban residential from 600 to 2,000 µg/g, and urban industrial from 900 to 16,000 µg/g. Brunekreef (1983) summarized studies of simultaneous measurements of air lead, soil lead, and household dust lead. With some exceptions, the household dust lead concentrations ranged from 400 to 700 µg/g per 1 µg/m^3 of lead in air. The relationship between household dust and soil dust was much broader. Because of the diverse nature of the studies, care should be taken in extrapolating these observations to more general circumstances. In El Paso, Texas, lead in household dust ranged from 2,800 to 100,000 µg/g within 2 km of a smelter (Landrigan et al.

1975). Davies et al. (1985) found a correlation between soil dust and household dust in an old lead mining area of North Wales, Great Britain, where a tenfold increase in soil lead was associated with a twofold increase in household dust lead.

It appears that most of the values for lead in dust in nonurban household environments fall in the range of 50-500 µg/g. A mean value of 300 µg/g is assumed. The only natural lead in dust would be some fraction of that derived from soil lead. A value of 10 µg/g seems reasonable, since some of the soil lead is of atmospheric origin. Since very little paint lead is included in the baseline estimate, most of the remaining dust lead would be from the atmosphere. Table 7-18 summarizes these estimates of human exposure to dusts for children and adults. It assumes that children ingest about five times as much dust as adults, most of the excess being street dusts from sidewalks and playgrounds. Exposure of children to occupational lead would be through contaminated clothing brought home by parents. Most of this lead is of undetermined origin because no data exist on whether the source is dust similar to household dust or unusual dust from the grinding and milling activities of factories.

7.3.1.5 Summary of Baseline Human Exposure to Lead. The values derived or assumed in the preceeding sections are summarized in Table 7-19. These values represent only consumption, not absorption, of lead by the human body. The key question of what are the risks to human health from these baseline exposures is addressed in Chapter 13. The approach used here to evaluate potential human exposure is similar to that used by the National Academy of Sciences (1980) and the Nutrition Foundation (1982) in their assessments of the impact of lead in the human environment.

7.3.2 Additive Exposure Factors

There are many conditions, even in nonurban environments, where an individual may increase his lead exposure by choice, habit, or unavoidable circumstance. The following sections describe these conditions as separate exposures to be added as appropriate to the baseline of human exposure described above. Most of these additive exposures clearly derive from air or dust, while a few derive from water or food.

7.3.2.1 Living and Working Environments With Increased Lead Exposure. Ambient air lead concentrations are typically higher in an urban than a rural environment. This factor alone can contribute significantly to the potential lead exposure of Americans, through increases in inhaled air and consumed dust. Produce from urban gardens may also increase the daily consumption of lead. Some environmental exposures may not be related only to urban living, such as houses with interior lead paint or lead plumbing, residences near smelters or refineries, or family gardens grown on high-lead soils. Occupational exposures may also occur in an urban or rural setting. These exposures, whether primarily in the occupational environment or

TABLE 7-19. SUMMARY OF BASELINE HUMAN EXPOSURES TO LEAD
(μg/day)

		Soil				
Source	Total lead consumed	Natural lead consumed	Indirect atmospheric lead*	Direct atmospheric lead*	Lead from solder or other metals	Lead of undetermined origin
Child-2 yr old						
Inhaled air	0.5	0.001	-	0.5	-	-
Food, Water & beverages	25.1	0.71	1.7	10.3	11.2	1.2
Dust	21.0	0.6	-	19.0	-	1.4
Total	46.6	1.3	1.7	29.8	11.2	2.6
Percent	100%	2.8%	3.5%	64.0%	24.0%	5.6%
Adult female						
Inhaled air	1.0	0.002	-	1.0	-	-
Food, Water & beverages	32.0	0.91	2.4	12.6	8.2	1.5
Dust	4.5	0.2	-	2.9	-	1.4
Total	37.5	1.2	2.5	17.4	13.5	2.9
Percent	100%	3.1%	6.6%	46.5%	36.1%	7.8%
Adult male						
Inhaled air	1.0	0.002	-	1.0	-	-
Food, Water & beverages	45.2	1.42	3.5	19.3	18.9	2.2
Dust	4.5	0.2	-	2.9	-	1.4
Total	50.7	1.6	3.5	23.2	18.9	3.6
Percent	100%	3.1%	6.8%	45.8%	37.2%	7.0%

*Indirect atmospheric lead has been previously incorporated into soil, and will probably remain in the soil for decades or longer. Direct atmospheric lead has been deposited on the surfaces of vegetation and living areas or incorporated during food processing prior to human consumption.

Source: This report.

secondarily in the home of the worker, would be additive with other exposures in an urban location or with special cases of lead-based paint or plumbing.

7.3.2.1.1 Urban atmospheres. Urban atmospheres have more airborne lead than do nonurban atmospheres, therefore there are increased amounts of lead in urban household and street dust. Typical urban atmospheres contain 0.5-1.0 µg Pb/m^3. Other variables are the amount of indoor filtered air breathed by urban residents, the amount of time spent indoors, and the amount of time spent on freeways. Reported means of urban dusts range from 500 to 3000 µg Pb/g. It is not known whether there is more or less dust in urban households and playgrounds than in rural environments. Whereas people may breathe the same amount of air, or eat and drink the same amount of food and water, it is not certain that urban residents consume the same amount of dust as nonurban. Nevertheless, in the absence of more reliable data, it has been assumed that urban and nonurban residents consume the same amount of dusts.

The indoor/outdoor ratio of atmospheric lead for urban environments is about 0.8 (Table 7-6). Assuming 2 hours of exposure/day outdoors at a lead concentration of 0.75 µg/m^3, 20 hours indoors at 0.6 µg/m^3, and 2 hours in a high traffic density area at 5 µg/m^3, a weighted mean air exposure of 1.0 µg/m^3 appears to be typical of urban residents.

7.3.2.1.2 Houses with interior lead paint. In 1974, the Consumer Product Safety Commission collected household paint samples and analyzed them for lead content (National Academy of Sciences; National Research Council, 1976). Analysis of 489 samples showed that 8 percent of the oil-based paints and 1 percent of the water-based paints contained greater than 0.5 percent lead (5000 µg Pb/g paint, based on dried solids), which was the statutory limit at the time of the study. The current statutory limit for Federal construction is 0.06 percent. The greatest amounts of leaded paint are typically found in the kitchens, bathrooms, and bedrooms (Tyler, 1970; Laurer et al., 1973; Gilbert et al., 1979).

Some investigators have shown that flaking paint can cause elevated lead concentrations in nearby soil. For example, Hardy et al. (1971) measured soil lead levels of 2000 µg/g next to a barn in rural Massachusetts. A steady decrease in lead level with increasing distance from the barn was shown, reaching 60 µg/g at fifty feet from the barn. Ter Haar and Aronow (1974) reported elevated soil lead levels in Detroit near eighteen old wood frame houses painted with lead-based paint. The average soil lead level within two feet of a house was just over 2000 µg/g; the average concentration at ten feet was slightly more than 400 µg/g. The same authors reported smaller soil lead elevations in the vicinity of eighteen brick veneer houses in Detroit. Soil lead levels near painted barns located in rural areas were similar to urban soil lead concentrations near painted houses, suggesting the importance of leaded paint at both urban and rural locations. The baseline lead concentration for household dust of 300 µg/g was increased to 2000 µg/g for houses with interior lead-based paints. The additional 1700 µg/g would add 85 µg Pb/day to the potential exposure of a child (Table 7-20). This increase would occur in an urban or nonurban environment and would be in addition to the urban residential increase if the lead-based painted house were in an urban environment.

7.3.2.1.3 Family gardens. Several studies have shown potentially higher lead exposure through the consumption of home-grown produce from family gardens grown on high lead soils or near sources of atmospheric lead. Mielke et al. (1983) surveyed the lead content of urban garden soils in Baltimore, finding concentrations ranging from 1 to 10,900, with a median of 100 µg/g. The soil sample was a mixture through 20-30 cm of the soil profile. The values greater than 100 µg/g were concentrated near the center of the city. Kneip (1978) found elevated levels of lead in leafy vegetables, root crops, and garden fruits associated qualitatively with traffic density and soil lead. Spittler and Feder (1978) reported a linear correlation between soil lead (100-1650 µg/g) and lead in or on leafy or root vegetables. Preer et al. (1980) found a threefold increase in lead concentrations of leafy vegetables (from 6 to 16 µg/g) in the soil lead range from 150 to 2200 µg/g. Chaney et al. (1984) have reviewed the recent studies on lead in urban gardens. In none of these studies were the lowest soil lead concentrations in the normal range of 10-25 µg/g, nor were any lead concentrations reported for vegetables as low as those of Wolnik et al. (1983, 1985) (see Table 7-8).

In family gardens, lead may reach the edible portions of vegetables by deposition of atmospheric lead directly on aboveground plant parts or on soil, or by the flaking of lead-containing paint chips from houses. Traffic density and distance from the road are not good predictors of soil or vegetable lead concentrations (Preer et al., 1980). Air concentrations and particle size distributions are the important determinants of deposition on soil or vegetation surfaces. Even at relatively high air concentrations (1.5 µg/m^3) and deposition velocity (0.5 cm/sec) (see Section 6.4.1), it is unlikely that surface deposition alone can account for more than 2-5 µg/g lead on the surface of lettuce during a 21-day growing period. It appears that a significant fraction of the lead in both leafy and root vegetables derives from the soil.

Using the same air concentration and deposition velocity values, a maximum of 1000 µg lead has been added to each cm^2 of the surface of the soil over the past 40 years. With cultivation to a depth of 15 cm, it is not likely that atmospheric lead alone can account for more than a few hundred µg/g of soil in urban gardens. Urban soils with lead concentrations of 500 µg/g or more must certainly have another source of lead. In the absence of a nearby (<5 km) stationary industrial source, paint chips seem the most likely explanation. Even if the house no longer stands at the site, the lead from paint chips may still be present in the soil.

Studies of family gardens do not agree on the concentrations of lead in produce. At the higher soil concentrations, Kneip (1978) reported 0.2-1 µg/g in vegetables, Spittler and Feder

TABLE 7-20. SUMMARY OF POTENTIAL ADDITIVE EXPOSURES TO LEAD
(μg/day)

Exposure	Total lead consumed	Atmospheric lead consumed	Other lead sources
Baseline exposure:			
Child			
Inhaled air	0.5	0.5	-
Food, water & beverages	25.1	10.3	14.8
Dust	21.0	19.0	2.0
Total baseline	46.6	29.8	16.8
Additional exposure due to:			
Urban atmospheres[1]	91	91	
Family gardens[2]	48	12	36
Interior lead paint[3]	110		110
Residence near smelter[4]	880	880	
Secondary occupational[5]	150		
Baseline exposure:			
Adult male			
Inhaled air	1.0	1.0	-
Food, water & beverages	54.7	20.3	34.4
Dust	4.5	2.9	1.6
Total baseline	60.2	24.2	36.0
Additional exposure due to:			
Urban atmospheres[1]	28	28	
Family gardens[2]	120	30	17
Interior lead paint[3]	17		
Residence near smelter[4]	100	100	
Occupational[6]	1100	1100	
Secondary occupational[5]	44		
Smoking[7]	30	27	3
Wine consumption[8]	100	?	?

[1] Includes lead from household (1000 μg/g) and street dust (1500 μg/g) and inhaled air (0.75 μg/m^3).

[2] Assumes soil lead concentration of 2000 μg/g; all fresh leafy and root vegetables, and sweet corn of Table 7-12 replaced by produce from garden. Also assumes 25% of soil lead is of atmospheric origin.

[3] Assumes household dust rises from 300 to 2000 μg/g. Dust consumption remains the same as baseline.

[4] Assumes household and street dust increase to 10,000 μg/g.

[5] Assumes household dust increases to 2400 μg/g.

[6] Assumes 8-hr shift at 10 μg Pb/m^3 or 90% efficiency of respirators at 100 μg Pb/m^3, and occupational dusts at 100,000 μg/m^3.

[7] One and a half packs per day.

[8] Assumes unusually high consumption of one liter per day.

(1978) reported 0.8-4.5 μg/g, and Preer et al. (1980) found 0.1-0.8 μg/g (all values converted to fresh weight). Since the Spittler and Feder (1978) and Preer et al. (1980) studies dealt with soils of about 2000 μg/g, these data can be used to calculate a worst case exposure of lead from family gardens. Assuming 0.8 μg/g for the leafy and root vegetables [compared to 0.01-0.05 μg/g of the Wolnik et al. (1983, 1985) study] family gardens could add 100 μg/day if the 137 g of leafy and root vegetables, sweet corn and potatoes consumed by adult males (Table 7D-1) were replaced by family garden products. Comparable values for children and adult females would be 40 and 80 μg/day, respectively. No conclusive data are available for vine vegetables, but the ranges of 0.8 to 0.1 μg/g for tomatoes suggest that the contamination by lead from soil is much less for vine vegetables than for leafy or root vegetables. Chaney et al. (1984) recommended that special precautions (extra washing and peeling) be taken with produce from urban gardens with soil lead from 500 to 1000 μg/g. They also recommended that leafy and root vegetables not be grown in gardens over 3000 μg/g.

7.3.2.1.4 Houses with lead plumbing. The Glasgow Duplicate Diet Study (United Kingdom Department of the Environment, 1982) reports that children approximately 13 weeks old living in houses with lead plumbing consume 6-480 μg Pb/day. Concentrations of lead in water ranged from less than 50 to over 500 μg/l for the 131 homes studied. Those children and mothers living in the homes containing high water-lead concentrations generally had greater total lead consumption and higher blood lead levels, according to the study. Breast-fed infants were exposed to much less lead than bottle-fed infants. Because the project was designed to investigate child and maternal blood lead levels over a wide range of water lead concentrations, the individuals studied do not represent a typical cross-section of the population. However, results of the study suggest that infants living in homes with lead plumbing may have exposure to considerable amounts of lead. This conclusion was also demonstrated by Sherlock et al. (1982) in a duplicate diet study in Ayr, Scotland.

7.3.2.1.5 Residences near smelters and refineries. Air lead concentrations within 2 km of lead smelters and refineries average 5-15 μg/m^3. Assuming the same indoor/outdoor ratio of atmospheric lead for nonurban residents (0.5), residents near smelters would be exposed to inhaled air lead concentrations of about 6 μg/m^3, compared to 0.05 μg/m^3 for the background levels. Household dust concentrations at El Paso, TX range from 3000 to 100,000 μg/g in 1982 (Landrigan et al., 1975). Morse et al. (1979) found that, with this installation of engineering improvements and pollution control, the dust lead was reduced to 1500-2000 by 1977. A value of 10,000 μg/g is assumed for household dust near a smelter. Between inhaled air and dust, a child in this circumstance would be exposed to 900 μg Pb/day above background levels. Exposures for adults would be much less, since they consume only 20 percent of the dusts children consume.

7.3.2.1.6 Occupational exposures. The highest and most prolonged exposures to lead are found among workers in the lead smelting, refining, and manufacturing industries (World Health Organization, 1977). In all work areas, the major route of lead exposure is by inhalation and ingestion of lead-bearing dusts and fumes. Airborne dusts settle out of the air onto food, water, the workers' clothing, and other objects, and may be transferred subsequently to the mouth. Therefore, good housekeeping and good ventilation have a major impact on exposure. It has been found that concentrations might be quite high in one factory and low in another solely because of differences in ventilation, or differences in custodial practices and worker education. The estimate of additional exposure in Table 7-20 is for an 8-hour shift at 100 μg Pb/m^3. Occupational exposure under these conditions is primarily determined by occupational dust consumed. Even tiny amounts (e.g., 10 mg) of dust containing 100,000 μg Pb/g dust can account for 1,000 μg/day exposure.

7.3.2.1.6.1 Lead mining, smelting, and refining. Roy (1977) studied exposures during mining and grinding of lead sulfide at a mill in the Missouri lead belt. Primary smelting operations were 4 km from the mill, hence the influence of the smelter was believed to be negligible. The total airborne lead levels were much greater than the concentrations of respirable lead, indicating a predominance of coarse material.

The greatest potential for high-level exposure exists in the process of lead smelting and refining (World Health Organization, 1977). The most hazardous operations are those in which molten lead and lead alloys are brought to high temperatures, resulting in the vaporization of lead. This is because condensed lead vapor or fume has, to a substantial degree, a small (respirable) particle size range. Although the total air lead concentration may be greater in the vicinity of ore-proportioning bins than it is in the vicinity of a blast furnace in a smelter, the amount of particle mass in the respirable size range may be much greater near the furnace.

A measure of the potential lead exposure in smelters was obtained in a study of three typical installations in Utah (World Health Organization, 1977). Air lead concentrations near all major operations, as determined using personal monitors worn by workers, were found to vary from about 100 to more than 4000 μg/m^3. Obviously, the hazard to these workers would be extremely serious if it were not for the fact that the use of respirators is mandatory in these particular smelters. Maximum airborne lead concentrations of about 300 μg/m^3 were measured in a primary lead-zinc smelter in the United Kingdom (King et al., 1979). These authors found poor correlations between airborne lead and blood lead in the smelter workers, and concluded that a program designed to protect these workers should focus on monitoring of biological parameters rather than environmental concentrations.

Spivey et al. (1979) studied a secondary smelter in southern California that recovers lead mainly from automotive storage batteries. Airborne lead concentrations of 10-4800 μg/m^3

were measured. The project also involved measurement of biological parameters as well as a survey of symptoms commonly associated with lead exposure; a poor correlation was found between indices of lead absorption and symptom reporting. The authors suggested that such factors as educational level, knowledge of possible symptoms, and biological susceptibility may be important factors in influencing symptom reporting. In a second article covering this same study, Brown et al. (1980) reported that smokers working at a smelter had greater blood lead concentrations than nonsmokers. Furthermore, smokers who brought their cigarettes into the workplace had greater blood lead concentrations than those who left their cigarettes elsewhere. It was concluded that direct environmental contamination of the cigarettes by lead-containing dust may be a major exposure pathway for these individuals (See Section 7.3.2.3.1).

Secondary lead smelters in Memphis, Tennessee and Salt Lake City, Utah were studied by Baker et al. (1979). The Memphis plant extracted lead principally from automotive batteries, producing 11,500 metric tons of lead in the eleven months preceding the measurements. The Salt Lake City plant used scrap to recover 258 metric tons of lead in the six months preceding the measurements. Airborne concentrations of lead in the Tennessee study exceeded 200 µg/m^3 in some instances, with personal air sampler data ranging from 120 µg/m^3 for a battery wrecker to 350 µg/m^3 for two yard workers. At the Utah plant, airborne lead levels in the office, lunchroom, and furnace room (furnace not operating) were 60, 90, and 100 µg/m^3, respectively. When charging the furnace, this value increased to 2650 µg/m^3. Personal samplers yielded concentrations of 17 µg/m^3 for an office worker, 700 µg/m^3 for two welders, and 2660 µg/m^3 for two furnace workers. Some workers in both plants showed clinical manifestations of lead poisoning; a significant correlation was found between blood lead concentrations and symptom reporting.

High levels of atmospheric lead are also reported in foundries in which molten lead is alloyed with other metals. Berg and Zenz (1967) found in one such operation that average concentrations of lead in various work areas were 280 to 600 µg/m^3. These levels were subsequently reduced to 30 to 40 µg/m^3 with the installation of forced ventilation systems to exhaust the work area atmosphere to the outside.

7.3.2.1.6.2 Welding and cutting of metals containing lead. When metals that contain lead or are protected with a lead-containing coating are heated in the process of welding or cutting, copious quantities of lead in the respirable size range may be emitted. Under conditions of poor ventilation, electric arc welding of zinc silicate-coated steel (containing 4.5 mg Pb/cm^2 of coating) produced breathing-zone concentrations of lead reaching 15,000 µg/m^3, far in excess of 450 µg/m^3, which is the current occupational short-term exposure limit (STEL) in the United States (Pegues, 1960). Under good ventilation conditions, a concentration of 140 µg/m^3 was measured (Tabershaw et al., 1943).

In a study of salvage workers using oxyacetylene cutting torches on lead-painted structural steel under conditions of good ventilation, breathing-zone concentrations of lead averaged 1200 μg/m^3 and ranged as high as 2400 μg/m^3 (Rieke, 1969). Lead poisoning in workers dismantling a painted bridge has been reported by Graben et al. (1978). Fischbein et al. (1978) discuss the exposure of workers dismantling an elevated subway line in New York City, where the lead content of the paint was as great as 40 percent. The authors report that one m^3 of air can contain 0.05 g lead at the source of emission. Similarly, Grandjean and Kon (1981) report elevated lead exposures of welders and other employees in a Baltimore, Maryland shipyard.

7.3.2.1.6.3 Storage battery industry. At all stages in battery manufacture except for final assembly and finishing, workers are exposed to high air lead concentrations, particularly lead oxide dust. For example, Boscolo et al. (1978) report air lead concentrations of 16-100 μg/m^3 in a battery factory in Italy, while values up to 1315 μg/m^3 have been measured by Richter et al. (1979) in an Israeli battery factory. Excessive concentrations, as great as 5400 μg/m^3, have been reported by the World Health Organization (1977).

7.3.2.1.6.4 Printing industry. The use of lead in typesetting machines has declined in recent years (see Table 5-1). Air concentrations of 10 to 30 μg/m^3 have been reported where this technique is used (Parikh et al., 1979). Lead is also a component of inks and dyes used in the printing industry, and consequently can present a hazard to workers handling these products.

7.3.2.1.6.5 Alkyl lead manufacture. Workers involved in the manufacture of alkyl lead compounds are exposed to both inorganic and alkyl lead. Some exposure also occurs at the petroleum refineries where the two compounds are blended into gasoline, but no data are available on these blenders.

The major potential hazard in the manufacture of tetraethyl lead and tetramethyl lead is from skin absorption, which is minimized by the use of protective clothing. Linch et al. (1970) found a correlation between an index of organic plus inorganic lead concentrations in a plant and the rate of lead excretion in the urine of workers. Significant concentrations of organic lead in the urine were found in workers involved with both tetramethyl lead and tetraethyl lead; lead levels in the tetramethyl lead workers were slightly higher because the reaction between the organic reagent and lead alloy takes place at a somewhat higher temperature and pressure than that employed in tetraethyl lead production.

Cope et al. (1979) used personal air samplers to assess exposures of five alkyl lead workers exposed primarily to tetraethyl lead. Blood and urine levels were measured over a six-week period. Alkyl lead levels in air ranged from 1.3 to 1249 μg/m^3, while inorganic lead varied from 1.3 to 62.6 μg/m^3. There was no significant correlation between airborne lead (either alkyl or inorganic) and blood or urine levels. The authors concluded that biological monitoring, rather than airborne lead monitoring, is a more reliable indicator of potential exposure problems.

7.3.2.1.6.6 Other occupations. In the rubber products industry and the plastics industry there are potentially high exposures to lead. The potential hazard of the use of lead stearate as a stabilizer in the manufacture of polyvinyl chloride was noted in the 1971 Annual Report of the British Chief Inspector of Factories (United Kingdom Department of Employment, Chief Inspector of Factories 1972). The inspector stated that the number of reported cases of lead poisoning in the plastics industry was second only to that in the lead smelting industry. Scarlato et al. (1969) reported other individual cases of exposure. The source of this problem is the dust that is generated when the lead stearate is milled and mixed with the polyvinyl chloride and the plasticizer. An encapsulated stabilizer which greatly reduces the occupational hazard is reported by Fischbein et al. (1982).

Sakurai et al. (1974), in a study of bioindicators of lead exposure, found ambient air concentrations averaging 58 μg/m^3 in the lead-covering department of a rubber hose manufacturing plant. Unfortunately, no ambient air measurements were taken for other departments or the control group.

The manufacture of cans with leaded seams may expose workers to elevated ambient lead levels. Bishop (1980) reports airborne lead concentrations of 25-800 μg/m^3 in several can manufacturing plants in the United Kingdom. Between 23 and 54 percent of the airborne lead was associated with respirable particles, based on cyclone sampler data.

Firing ranges may be characterized by high airborne lead concentrations, hence instructors who spend considerable amounts of time in such areas may be exposed to lead. For example, Smith (1976) reports airborne lead concentrations of 30-160 μg/m^3 at a firing range in the United Kingdom. Anderson et al. (1977) discuss lead poisoning in a 17 year old male employee of a New York City firing range, where airborne lead concentrations as great as 1000 μg/m^3 were measured during sweeping operations. Another report from the same research group presents time-weighted average exposures of instructors of 45-900 μg/m^3 in three New York City firing ranges (Fischbein et al., 1979).

Removal of leaded paint from walls and other surfaces in old houses may pose a health hazard. Feldman (1978) reports an airborne lead concentration of 510 μg/m^3, after 22 minutes of sanding an outdoor post coated with paint containing 2.5 mg Pb/cm^2. After only five minutes of sanding an indoor window sill containing 0.8-0.9 mg Pb/cm^2, the air contained 550 μg/m^3. Homeowners who attempt to remove leaded paint themselves may be at risk of excessive lead exposure. Garage mechanics may be exposed to excessive lead concentrations. Clausen and Rastogi (1977) report airborne lead levels of 0.2-35.5 μg/m^3 in ten garages in Denmark; the greatest concentration was measured in a paint workshop. Used motor oils were found to contain 1500-3500 μg Pb/g, while one brand of unused gear oil contained 9280 μg Pb/g. The authors state that absorption through damaged skin could be an important exposure pathway. Other occupations involving risk of lead exposure include stained glass manufacturing and repair, arts and crafts, and soldering and splicing.

7.3.2.1.7 Secondary occupational exposure. Winegar et al. (1977) examined environmental concentrations as well as biological indicators and symptom reporting in workers in a secondary lead smelter near St. Paul, Minnesota. The smelter recovers approximately 9000 metric tons of lead per year from automotive batteries. The lead concentrations in cuff dust from trousers worn by two workers were 60,000 and 600,000 μg/g. The amount of lead contained in pieces of cloth 1 cm^2 cut from the bottoms of trousers worn by the workers ranged from 110 to 3000 μg, with a median of 410 μg. In all cases, the trousers were worn under coveralls. Dust samples from 25 households of smelter workers ranged from 120 to 26,000 μg/g, with a median of 2400 μg/g. No significant correlations were found between dust lead concentrations and biological indicators, or between symptom reporting and biological indicators. However, there was an increased frequency of certain objective physical signs, possibly due to lead toxicity, with increased blood lead level. The authors also concluded that the high dust lead levels in the workers' homes are most likely due to lead originating in the smelter.

7.3.2.2 Additive Exposure Due to Age, Sex, or Socio-Economic Status

7.3.2.2.1 Quality and quantity of food. The quantity of food consumed per body weight varies greatly with age and somewhat with sex. A 14 kg, 2-year-old child eats and drinks 1.5 kg food and water per day. This is 110 g/kg, or 3 times the consumption of an 80 kg adult male, who eats 39 g/kg. Teenage girls consume less than boys and elderly women eat more than men, on a body weight basis.

It is likely that poor people eat more canned foods and less frozen and pre-prepared foods. Rural populations probably eat more home-grown foods and meats packed locally.

7.3.2.2.2 Mouthing behavior of children. Children place their mouths on dust-collecting surfaces and lick non-food items with their tongues. This fingersucking and mouthing activity are natural forms of behavior for young children that expose them to some of the highest concentrations of lead in their environment. A single gram of dust may contain ten times more lead than the total diet of the child.

7.3.2.3 Special Habits or Activities.

Rabinowitz and Needleman (1984) found a positive correlation between cord blood lead and such maternal exposure factors as use of tobacco, hard alcohol, coffee, and amount of lead in dust. Factors unrelated to cord blood lead levels were amount of dust, tap water lead, air lead, and lead paint. One or more of the above exposure factors may be correlated with other factors, such as race, marital status, schooling, or maternal age. Of these, race and marital status demonstrated a relationship to blood lead. Whereas this study did not attempt to quantify actual exposure, it does identify several activities that are likely to increase human exposure to lead.

7.3.2.3.1 Smoking. Lead is also present in tobacco. The World Health Organization (1977) estimates a lead content of 2.5-12.2 μg per cigarette; roughly two to six percent of this lead may be inhaled by the smoker. The National Academy of Sciences (1980) has used these data to conclude that a typical urban resident who smokes 30 cigarettes per day may inhale roughly equal amounts of lead from smoking and from breathing urban air.

7.3.2.3.2 Alcoholic beverages. Reports of lead in European wines (Olsen et al., 1981; Boudene et al., 1975; Zurlo and Griffini, 1973) show concentrations averaging 100-200 μg/l and ranging as high as 300 μg/l. Measurements of lead in domestic wines were in the range of 100-300 μg/l for California wines with and without lead foil caps. The U.S. Food and Drug Administration (1983) found 30 μg/l in the 1982 Market Basket Survey. The average daily consumption of table wine by a 25- to 30-year-old adult in the U.S. is about 12 g. Even with a lead content of 0.1 μg/g, which is ten times higher than drinking water, wine does not appear to represent a significant potential exposure to lead. At one liter per day, however, lead consumption in wine would be greater than the total baseline consumption.

McDonald (1981) points out that older wines with lead foil caps may represent a hazard, especially if they have been damaged or corroded. Wai et al. (1979) found that the lead content of wine rose from 200 to 1200 μg/l when the wine was allowed to pass over the thin ring of residue left by the corroded lead foil cap. Newer wines (1971 and later) use other means of sealing. If a lead foil is used, the foil is tin-plated and coated with an acid-resistant substance. Lead concentrations in beer are generally lower than those in wine; Thalacker (1980) reports a maximum concentration of 80 μg/l in several brands of German beer. The U.S. Food and Drug Administration (1983) found 13 μg/l in beer consumed by Americans (Table 7D-1).

7.3.2.3.3 Pica. Pica is the compulsive, habitual consumption of non-food items, such as paint chips and soil. This habit can present a significant lead exposure to the afflicted person, especially to children, who are more apt to have pica. There are very little data on the amounts of paint or soil eaten by children with varying degrees of pica. Exposure can only be expressed on a unit basis. Billick and Gray (1978) report lead concentrations of 1000 to 5000 μg/cm^2 surface area in lead-based paint pigments. To a child with pica, a single chip of paint can represent greater exposure than any other source of lead. A gram of urban soil may have 150 to 2000 μg lead.

7.3.2.3.4 Glazed earthenware vessels. Another potential source of dietary lead poisoning is the use of inadequately glazed earthenware vessels for food storage and cooking. An example of this danger involved the severe poisoning of a family in Idaho that resulted from drinking orange juice that had been stored in an earthenware pitcher (Block, 1969). Similar cases, sometimes including fatalities, have involved other relatively acidic beverages such as fruit juices and soft drinks, and have been documented by other workers (Klein et al., 1970; Harris and Elsen, 1967). Because of these incidents, the U.S. Food and Drug Administration (1980) has established a maximum permissible concentration of 2.5-7 μg Pb/ml in solution after leaching with 4 percent acetic acid in the kitchenware for 24 hours, depending on the shape and volume of the vessel.

Inadequately glazed pottery manufactured in other countries continues to pose a significant health hazard. For example, Spielholtz and Kaplan (1980) report 24-hour acetic acid-leached lead concentrations as great as 4400 μg/g in Mexican pottery. The leached lead decreased with exposure time, and after several days appears to asymptotically approach a value which may be as high as 600 μg/g. These investigators have also measured excessive lead concentrations leached into acidic foods cooked for two hours in the same pottery. Similarly, Acra et al. (1981) report that 85 percent of 275 earthenware vessels produced in primitive Lebanese potteries had lead concentrations above the 7 μg/g limit set by the U.S. FDA. However, only 9 percent of 75 vessels produced in a modern Beirut pottery exceeded the limit. Cubbon et al. (1981) have examined properly glazed ceramic plates in the United Kingdom, and have found a decrease in leached lead with exposure time down to very low levels. The authors state that earthenware satisfying the 7 μg/g limit can contribute about 3 μg/day to the dietary intake of the average consumer.

7.3.2.3.5 Hobbies. There are a few hobbies where the use of metallic lead or solder may present a hazard to the user. Examples are electronics projects, stained glass window construction, and firing range ammunition recovery. There are no reports in which the exposure to lead has been quantified during these activities.

7.3.3 Summary of Additive Exposure Factors

Beyond the baseline level of human exposure, additional amounts of lead consumption are largely a matter of individual choice or circumstance. Many of these additional exposures arise from the ingestion of atmospheric lead in dust. In one or more ways probably 90 percent of the American population are exposed to lead at greater than baseline levels. A summary of the most common additive exposure factors appears in Table 7-20. In some cases, the additive exposure can be fully quantified and the amount of lead consumed can be added to the baseline consumption. These may be continuous (urban residence), or seasonal (family gardening) exposures. Some factors can be quantified only on a unit basis because of wide ranges in exposure duration or concentration. For example, factors affecting occupational exposure are air lead concentrations (10-4000 μg/m^3), use and efficiency of respirators, length of time of exposure, dust control techniques, and worker training in occupational hygiene.

7.4 SUMMARY

Ambient airborne lead concentrations have shown no marked trend from 1965 to 1977. Over the past five years, however, distinct decreases have occurred. The mean air concentration has dropped from 1.3 $\mu g/m^3$ in 1977 to 0.40 $\mu g/m^3$ in 1984. This decrease reflects the lower lead emissions from mobile sources in recent years. Airborne size distribution data indicate that most of the airborne lead mass is found in submicron particles.

Atmospheric lead is deposited on vegetation and soil surfaces, entering the human food chain through contamination of grains and leafy vegetables, of pasture lands, and of soil moisture taken up by all crops. Lead contamination of drinking water supplies appears to originate mostly from within the distribution system.

Most people receive the largest portion of their lead intake through foods. Unprocessed foods such as fresh fruits and vegetables receive lead by atmospheric deposition as well as uptake from soil; crops grown near heavily traveled roads generally have greater lead levels than those grown at greater distances from traffic. For many crops the edible internal portions of the plant (e.g., kernels of corn and wheat) have considerably less lead than the outer, more exposed parts, such as stems, leaves, and husks. Atmospheric lead accounts for about 45 percent of the total adult lead exposure, and 65 percent of the exposure for children. Processed foods have greater lead concentrations than unprocessed foods, due to lead inadvertently added during processing. Foods packaged in soldered cans have much greater lead levels than foods packaged in other types of containers. About 35 percent of the baseline adult exposure to lead results from the use of solder lead in packaging food and distributing drinking water.

Significant amounts of lead in drinking water can result from contamination at the water source and from the use of lead solder in the water distribution system. Atmospheric deposition has been shown to increase lead in rivers, reservoirs, and other sources of drinking water; in some areas, however, lead pipes pose a more serious problem. Soft, acidic water in homes with lead plumbing may have excessive lead concentrations. Besides direct consumption of the water, exposure may occur when vegetables and other foods are cooked in water containing lead.

All of the categories of potential lead exposure discussed above may influence or be influenced by dust and soil. For example, lead in street dust is derived primarily from vehicular emissions, while in house dust may originate from nearby stationary or mobile sources. Food and water may include lead adsorbed from soil as well as deposited atmospheric material. Flaking lead-based paint has been shown to increase soil lead levels. Natural concentrations of lead in soil average approximately 15 $\mu g/g$; this natural lead, in addition to anthropogenic lead emissions, influences human exposure.

Americans living in rural areas away from sources of atmospheric lead consume 35-55 µg Pb/day from all sources. Circumstances that can increase this exposure are urban residence (25-100 µg/day), family garden on high-lead soil (40-100 µg/day), houses with interior lead-based paint (20-85 µg/day), and residence near a smelter (400-900 µg/day). Occupational settings, smoking, and wine consumption also can increase consumption of lead according to the degree of exposure.

A number of manmade materials are known to contain lead, the most important being paint and plastics. Lead-based interior paints, although no longer used, are a major problem in older homes. Small children who ingest paint flakes can receive excessive lead exposure. Incineration of plastics may emit large amounts of lead into the atmosphere. Because of the increasing use of plastics, this source is likely to become more important. Other manmade materials containing lead include colored dyes, cosmetic products, candle wicks, and products made of pewter and silver.

The greatest occupational exposures are found in the lead smelting and refining industries. Excessive airborne lead concentrations and dust lead levels are occasionally found in primary and secondary smelters; smaller exposures are associated with mining and processing of the lead ores. Welding and cutting of metal surfaces coated with lead-based paint may also result in excessive exposure. Other occupations with potentially high exposures to lead include the manufacture of lead storage batteries, printing equipment, alkyl lead, rubber products, plastics, and cans; individuals removing lead paint from walls and those who work in indoor firing ranges may also be exposed to lead.

Environmental contamination by lead should be measured in terms of the total amount of lead emitted to the biosphere. American industry contributes several hundred thousand tons of lead to the environment each year: 55,000 tons from petroleum additives, 50,000 tons from ammunition, 45,000 tons in glass and ceramic products, 16,000 tons in paint pigments, 8,000 tons in food can solder, and untold thousands of tons of captured wastes during smelting, refining, and coal combustion. These are uses of lead that are generally not recoverable, thus they represent a permanent contamination of the human or natural environment. Although much of this lead is confined to municipal and industrial waste dumps, a large amount is emitted to the atmosphere, waterways, and soil, to become a part of the biosphere.

Potential human exposure can be expressed as the concentrations of lead in these environmental components (air, dust, food, and water) that interface with man. It appears that, with the exception of extraordinary cases of exposure, about 80 to 100 µg of lead are consumed daily by each American, including additional exposure above baseline.

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APPENDIX 7A
SUPPLEMENTAL AIR MONITORING INFORMATION

7A.1 AIRBORNE LEAD SIZE DISTRIBUTION

In Section 7.2.1.3.1, several studies of the particle size distributions for atmospheric lead were discussed. The distributions at forty locations were given in Figure 7-5. Supplementary information from each of these studies is given in Table 7A-1.

TABLE 7A-1. INFORMATION ASSOCIATED WITH THE AIRBORNE LEAD SIZE DISTRIBUTIONS OF FIGURE 7-5

Graph no.	Reference	Dates of sampling	Location of sampling	Type of sampler	C_T µg/m³	Approx. MMD µm
1	Lee et al. (1972)	Jan. - Dec. 1970 Average of 4 quarterly composited samples, representing a total of 21 sampling periods of 24 hours each	Chicago, Illinois	Modified Anderson impactor with backup filter	3.2	0.68
2	Lee et al. (1972)	Mar. - Dec. 1970 Same averaging as Graph 1, total of 18 sampling periods	Cincinnati, Ohio	Modified Andersen impactor with backup filter	1.8	0.48
3	Lee et al. (1972)	Jan. - Dec. 1970 Same averaging as Graph 1, total of 21 sampling periods	Denver, Colorado	Modified Andersen impactor with backup filter	1.8	0.50
4	Lee et al. (1972)	Mar. - Dec. 1970 Same averaging as Graph 1, total of 20 sampline periods	Philadelphia, Pennsylvania	Modified Andersen impactor with backup filter	1.6	0.47
5	Lee et al. (1972)	Jan. - Dec. 1970 Same averaging as Graph 1, total of 22 sampling periods	St. Louis, Missouri	Modified Andersen impactor with backup filter	1.8	0.69
6	Lee et al. (1972)	Jan. - Dec. 1970 Same averaging as Graph 1, total of 23 sampling periods	Washington, D.C.	Modified Andersen impactor with backup filter	1.3	0.42

TABLE 7A-1. (continued)

Graph no	Reference	Dates of sampling	Location of sampling	Type of sampler	C_T µg/m³	Approx. MMD µm
7	Lee et al. (1968)	September 1966 Average of 14 runs, 24 hours each	Cincinnati, Ohio	Andersen impactor with backup filter, 1.2m above the ground	2.8	0.29
8	Lee et al. (1968)	February 1967 Average of 3 runs 4 days each	Fairfax, Ohio suburb of Cincinnati	Andersen impactor with backup filter, 1.2m above the ground	0.69	0.42
9	Peden (1977)	Summer 1975 Average of 4 runs, average 8 days each	Alton, Illinois, industrial area near St. Louis	Andersen impactor no backup filter	0.24	2.1
10	Peden (1977)	Summer 1972 Average of 3 runs, average 10 days each	Centreville, Illinois, downwind of a zinc smelter	Andersen impactor with backup filter	0.62	0.41
11	Peden (1977)	Summer 1973 Average of 2 runs average 5 days each	Collinsville, Illinois industrial area near St. Louis	Andersen impactor with backup filter	0.67	0.24
12	Peden (1977)	Summer 1973 Average of 2 runs, average 6 days each	KMOX radio transmitter, Illinois, industrial area near St. Louis	Andersen impactor with backup filter	0.60	0.31
13	Peden (1977)	Summer 1972 Average of 9 runs, average 9 days each	Pere Marquette State Park, Illionis, upwind of St. Louis	Andersen impactor with backup filter	0.15	0.51
14	Peden (1977)	Summer 1975 Average of 4 runs, average 8 days each	Wood River, Illinois, industrial area near St. Louis	Andersen impactor, no backup filter	0.27	1.8

TABLE 7A-1 (continued)

Graph no	Reference	Dates of sampling	Location of sampling	Type of sampler	C_T µg/m³	Approx. MMD µm
15	Cholak et al. (1968)	April 1968 average of several runs, 3 days each	3 sites: 10,400 and 3300m from Interstate 75, Cincinnati, Ohio	Andersen impactor with backup filter	7.8* 1.7 1.1	0.32
16	McDonald and Duncan (1979)	June 1975 One run of 15 days	Glasgow, Scotland	Casella impactor with backup filter, 30m above the ground	0.53	0.51
17	Dorn et al. (1976)	Winter, spring, summer 1972 Average of 3 runs, 27 days each	Southeast Missouri, 800m from a lead smelter	Andersen impactor, no backup filter, 1.7m above the ground	1.0	3.8
18	Dorn et al. (1976)	Winter, spring, summer 1972 Average of 3 runs, 14 days each	Southeast Missouri, 75 km from the lead smelter of Graph 17	Andersen impactor, no backup filter, 1.7m above the ground	0.11	2.4
19	Daines et al. (1970)	1968 Average of continuous 1-week runs over an 8-month period	3 sites: 9, 76, and 530m from U.S. Route 1, New Brunswick, New Jersey	Cascade impactor with backup filter	4.5 2.2 1.5	0.35
20	Martens et al. (1973)	July 1971 One run of 4 days	9 sites throughout San Francisco area	Andersen impactor with backup filter	0.84	0.49
21	Lundgren (1970)	November 1968 Average of 10 runs, 16 hours each	Riverside, California	Lundgren impactor	0.59	0.50
22	Huntzicker et al. (1975)	May 1973 One run of 8 hours	Shoulder of Pasadena Freeway near downtown Los Angeles, California	Andersen impactor with backup filter, 2m above the ground	14.0	0.32

TABLE 7A-1 (continued)

Graph no	Reference	Dates of sampling	Location of sampling	Type of sampler	C_T μg/m³	Approx. MMD μm
23	Huntzicker et al. (1975)	Februray 1974 One run of 6 days	Pasadena, California	Andersen impactor with backup filter, on roof of 4 story building	3.5	0.72
24	Davidson (1977)	May and July 1975 Average of 2 runs, 61 hours each	Pasadena, California	Modified Andersen impactor with backup filter on roof of 4 story building	1.2	0.97
25	Davidson et al. (1980)	October 1979 One run of 120 hours	Clingman's Dome Great Smokies National Park, elev. 2024m	2 Modified Andersen impactors with backup filters, 1.2m above the ground	0.014	1.0
26	Davidson et al. (1981a)	July-Sep. 1979 Average of 2 runs, 90 hours each	Pittsburgh, Pennsylvania	Modified Andersen impactor with backup filter, 4m above the ground	0.60	0.56
27	Davidson et al. (1981b)	December 1979 One run of 52 hours	Nepal Himalayas elev. 3962m	Modified Andersen impactor with backup filter, 1.2m above the ground	0.0014	0.54
28	Goold and Davidson (1982)	June 1980 One run of 72 hours	Export, Pennsylvania rural site 40 km east of Pittsburgh	2 Modified Andersen impactors with backup filters, 1.2m above the ground	0.111	1.2
29	Goold and Davidson (1982)	July 1980 One run of 34 hours	Packwood, Washington rural site in Gifford Pinchot National Forest	Modified Andersen impactor with backup filter, 1.5m above the ground	0.016	0.40

TABLE 7A-1 (continued)

Graph no	Reference	Dates of sampling	Location of sampling	Type of sampler	C_T µg/m^3	Approx. MMD µm
30	Goold and Davidson (1982)	July-Aug. 1980 One run of 92 hours	Hurricane Ridge Olympic National Park elev. 1600m	Modified Andersen impactor with backup filter, 1.5m above the ground	0.0024	0.87
31	Duce et al. (1976)	May - June 1975 One run of 112 hours	Southeast coast of Bermuda	Sierra high-volume impactor with backup filter, 20m above the ground	0.0085	0.57
32	Duce et al. (1976)	July 1975 One run of 79 hours	Southeast coast of Bermuda	Sierra high-volume impactor with backup filter, 20m above the ground	0.0041	0.43
33	Harrison et al. (1971)	April 1968 Average of 21 runs, 2 hours each	Ann Arbor, Michigan	Modified Andersen impactor with backup filter, 20m above the ground	1.8	0.16
34	Gillette and Winchester (1972)	Oct. 1968 Average of 15 runs, 24 hours each	Ann Arbor, Michigan	Andersen impactor with backup filter	0.82	0.28
35	Gillette and Winchester (1972)	May - Sept. 1968 Average of 10 runs, 8 hours each	Chicago, Illinois	Andersen impactor with backup filter	1.9	0.39
36	Gillette and Winchester (1972)	Oct. 1968 Average of 3 runs, 24 hours each	Lincoln, Nebraska	Andersen impactor with backup filter	0.14	0.42
37	Johansson et al. (1976)	June - July 1973 Average of 15 runs, average 50 hr each	2 sites in Tallahassee, Florida	Delron Battelle-type impactor, no backup filter, on building roofs	0.24	0.62

TABLE 7A-1 (continued)

Graph no	Reference	Dates of sampling	Location of sampling	Type of sampler	C_T µg/m³	Approx. MMD µm
38	Cawse et al. (1974)	July - Dec. 1973	Chilton, England	Andersen impactor with backup filter, 1.5m above the ground	0.16	0.57
39	Pattenden et al. (1974)	May - Aug. 1973 Average of 4 runs, 1 month each	Trebanos, England	Andersen impactor with backup filter, 1.5m above the ground	0.23	0.74
40	Bernstein and Rahn (1979)	Aug. 1976 Average of 4 runs, 1 week each	New York City	Cyclone sampling system with backup filter, on roof on 15 story building	1.2	0.64

*Airborne concentrations for filters run at the same sites as the impactor, but during different time periods. Impactor concentrations not available.

APPENDIX 7B
SUPPLEMENTAL SOIL AND DUST INFORMATION

Lead in soil, and dust of soil origin, is discussed in Section 7.2.2. The data show average soil concentrations are 8-25 µg/g, and dust from this soil rarely exceeds 80-100 µg/g. Street dust, household dust, and occupational dusts often exceed this level by one to two orders of magnitude. Tables 7B-1 and 7B-2 summarize several studies of street dust. Table 7B-3 shows data on household and residential soil dust. These data support the estimates of mean lead concentrations in dust discussed in Section 7.3.1.4. Table 7B-4 gives airborne lead concentrations for an occupational setting, which are only qualitatively related to dust lead concentrations.

TABLE 7B-1. LEAD DUST ON AND NEAR HEAVILY TRAVELED ROADWAYS

Sampling site	Concentration µg Pb/g	Reference
Washington, DC:		
Busy intersection	13,000	
Many sites	4000-8000	Fritsch and Prival (1972)
Chicago:		
Near expressway	6600	
Philadelphia:		
Near expressway	3000-8000	Kennedy (1973)
Brooklyn:		
Near expressway	900-4900	Lombardo (1973)
New York City:		
Near expressway	2000	Pinkerton et al. (1973)
Detroit:		
Street dust	970-1200	Ter Haar and Aronow (1974)
Philadelphia:		
Gutter (low pressure)	210-2600	Shapiro et al. (1973)
Gutter (high pressure)	280-8200	Shapiro et al. (1973)
Miscellaneous U.S. Cities:		
Highways and tunnels	10,000-20,000	Buckley et al. (1973)
Netherlands:		
Heavily traveled roads	5000	Rameau (1973)

TABLE 7B-2. LEAD CONCENTRATIONS IN STREET DUST IN LANCASTER, ENGLAND

Site	No. of samples	Range of concentrations	Mean	Standard deviation
Car parks	4	39,700 - 51,900	46,300	5,900
	16	950 - 15,000	4,560	3,700
Garage forecourts	2	44,100 - 48,900	46,500	--
	7	1,370 - 4,480	2,310	1,150
Town centre streets	13	840 - 4,530	2,130	960
Main roads	19	740 - 4,880	1,890	1,030
Residential areas	7	620 - 1,240	850	230
Rural roads	4	410 - 870	570	210

Source: Harrison (1979).

TABLE 7B-3. LEAD DUST IN RESIDENTIAL AREAS

Sampling site	Concentration (μg Pb/g)	Reference
Philadelphia:		
Classroom	2000	
Playground	3000	
Window frames	1750	Shapiro et al. (1973)
Boston and New York:		
House dust	1000-2000	Needleman and Scanlon (1973)
Brattleboro, VT:		
In home	500-900	Darrow and Schroeder (1974)
New York City:		
Middle Class Residential	610-740	Pinkerton et al. (1973)
Philadelphia:		
Urban industrial	930-16,000	Needleman et al. (1974)
Residential	290-1000	Needleman et al. (1974)
Suburban	280-1500	Needleman et al. (1974)
Derbyshire, England:		
Low soil lead area	130-3000	Barltrop et al. (1975)
High soil lead area	1050-28,000	Barltrop et al. (1975)

• TABLE 7B-4. AIRBORNE LEAD CONCENTRATIONS BASED ON PERSONAL SAMPLERS, WORN BY EMPLOYEES AT A LEAD MINING AND GRINDING OPERATION IN THE MISSOURI LEAD BELT (μg/m^3)

Occupation	N*	High	Low	Mean
Mill operator	6	300	50	180
Flotation operator	4	750	100	320
Filter operator	4	2450	380	1330
Crusher operator	4	590	20	190
Sample finisher	2	10,000	7070	8530
Crusher utility	1	--	--	70
Shift boss	5	560	110	290
Equipment operator	1	--	--	430

*N denotes number of air samples.

Source: Roy (1977).

APPENDIX 7C
STUDIES OF SPECIFIC POINT SOURCES OF LEAD

This collection of studies is intended to extend and detail the general picture of lead concentrations in proximity to identified major point sources as portrayed in Chapter 7. Because emissions and control technology vary between point sources, each point source is unique in the degree of environmental contamination. The list is by no means all-inclusive, but is intended to be representative and to supplement the data cited in Chapter 7. In many of the studies, blood samples of workers and their families were taken. These studies are also discussed in Chapter 11.

7C.1 SMELTERS AND MINES

7C.1.1 Two Smelter Study

The homes of workers of two unidentified secondary lead smelters in different geographical areas of the United States were studied by Rice et al. (1978). Paper towels were used to collect dust from surfaces in each house, following the method of Vostal et al. (1974). A total of 33 homes of smelter workers and 19 control homes located in the same or similar neighborhoods were investigated. The geometric mean lead levels on the towels were 79.3 μg (smelter workers) versus 28.8 μg (controls) in the first area, while in the second area mean values were 112 μg versus 9.7 μg. Also in the second area, settled dust above doorways was collected by brushing the dust into glassine envelopes for subsequent analysis. The geometric mean lead content of this dust in 15 workers' homes was 3300 μg/g, compared with 1200 μg/g in eight control homes. Curbside dust collected near each home in the second area had a geometric mean lead content of 1500 μg/g, with no significant difference between worker and control homes. No significant difference was reported in the paint lead content between worker and control homes. The authors concluded that lead in dust carried home by these workers contributed to the lead content of dust in their homes, despite showering and changing clothes at the plant, and despite work clothes being laundered by the company. Storage of employee street clothes in dusty lockers, walking across lead-contaminated areas on the way home, and particulate settling on workers' cars in the parking lot may have been important factors. Based on measurement of zinc protoporphyrin levels in the blood of children in these homes, the authors also concluded that the greater lead levels in housedust contributed to increased child absorption of lead.

7C.1.2 British Columbia, Canada

Neri et al. (1978) and Schmitt et al. (1979) examined environmental lead levels in the vicinity of a lead-zinc smelter at Trail, British Columbia. Total emissions from the smelter averaged about 135 kg Pb/day. Measurements were conducted in Trail (population 12,000), in Nelson, a control city 41 km north of Trail (population 10,000), and in Vancouver. The annual mean airborne lead concentrations in Trail and in Nelson were 2.0 and 0.5 μg/m^3, respectively. Mean lead levels in surface soil were 1320 μg/g in Trail (153 samples), 192 μg/g in Nelson (55 samples), and 1545 μg/g in Vancouver (37 samples).

Blood lead measurements show a positive correlation with soil lead levels for children aged 1-3 years and for first graders, but no significant correlation for ninth graders. The authors concluded that small children are most likely to ingest soil dust, and hence deposited smelter-emitted lead may pose a potential hazard for the youngest age group.

7C.1.3 Netherlands

Environmental lead concentrations were measured in 1978 near a secondary lead smelter in Arnhem, Netherlands (Diemel et al., 1981). Air and dust were sampled in over 100 houses at distances of 450-1000 meters from the smelter, with outdoor samples of air, dust, and soil collected for comparison. Results are presented in Table 7C-1. Note that the mean indoor concentration of total suspended particulates (TSP) is greater than the mean outdoor concentration, yet the mean indoor lead level is smaller than the corresponding outdoor level. The authors reasoned that indoor sources such as tobacco smoke, consumer products, and decay of furnishings are likely to be important in affecting indoor TSP; however, much of the indoor lead was probably carried in from the outside by the occupants, e.g., as dust adhering to shoes. The importance of resuspension of indoor particles by activity around the house was also discussed.

7C.1.4 Belgium

Roels et al. (1978, 1980) measured lead levels in the air, in dust, and on childrens' hands at varying distances from a lead smelter in Belgium (annual production 100,000 metric tons). Blood data from children living near the smelter were also obtained. Air samples were collected nearly continuously beginning in September 1973. Table 7C-2 lists the airborne concentrations recorded during five distinct population surveys between 1974 and 1978, while Figure 7C-1 presents air, dust, and hand data for Survey #3 in 1976. Statistical tests showed that blood lead levels were better correlated with lead on childrens' hands than with air lead. The authors suggested that ingestion of contaminated dust by hand-to-mouth activities

TABLE 7C-1. LEAD CONCENTRATIONS IN INDOOR AND OUTDOOR AIR, INDOOR AND OUTDOOR DUST, AND OUTDOOR SOIL NEAR THE ARNHEM, NETHERLANDS SECONDARY LEAD SMELTER (INDOOR CONCENTRATIONS)

Parameter	Arithmetic mean	Range	n*
Suspended particulate matter			
dust concentration (μg/m^3)	140	20-570	101
lead concentration (μg/m^3)	0.27	0.13-0.74	101
dust lead content (μg/kg)	2670	400-8200	106
Dustfall			
dust deposition (mg/m^2·day)	15.0	1.4-63.9	105
lead deposition (μg/m^2·day)	9.30	1.36-42.4	105
dust lead content (mg/kg)	1140	457-8100	105
Floor dust			
amount of dust (mg/m^2)	356	41-2320	107
amount of lead (μg/m^2)	166	18-886	101
Dust lead content (mg/kg)			
in "fine" floor dust	1050	463-4740	107
in "coarse" floor dust	370	117-5250	101

*N number of houses.

(OUTDOOR CONCENTRATIONS)

Parameter	Arithmetic mean	Range
Suspended particles		
dust concentration (μg/m^3)	64.5	53.7-73.3
lead concentraton (μg/m^3) (high-volume samplers, 24-hr samples, 2 month's average)	0.42	0.28-0.52
Lead in dustfall (μg/m^2·day) (deposit gauges, weekly samples, 2 months' average)	508	208-2210
Lead in soil (mg/kg 0-5 cm)	322	21-1130
Lead in streetdust (mg/kg <0.3 mm)	860	77-2670

Source: Diemel et al. (1981).

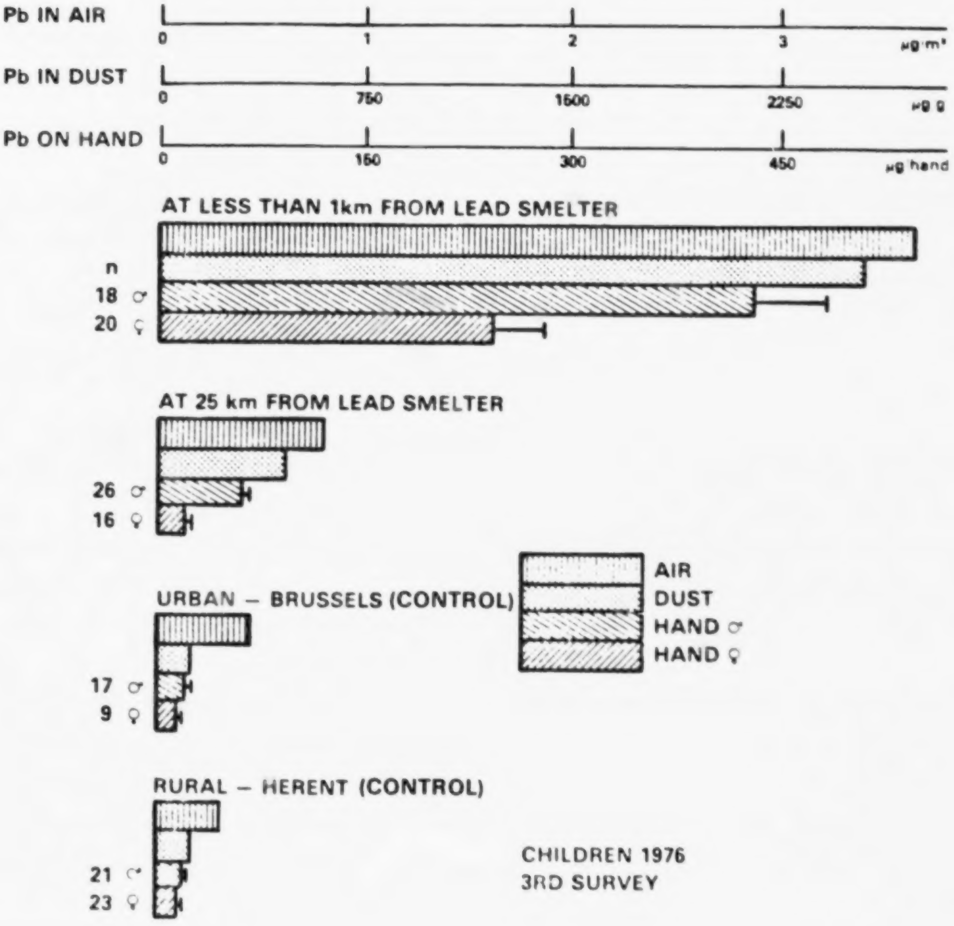

Figure 7C-1. Concentrations of lead in air, in dust, and on children's hands, measured during the third population survey of Table E. Values obtained less than 1 km from the smelter, at 2.5 km from the smelter, and in two control areas are shown. The number of children (n) is shown by sex.

Source: Roels et al. (1980).

TABLE 7C-2. AIRBORNE CONCENTRATIONS OF LEAD DURING FIVE POPULATION SURVEYS NEAR A LEAD SMELTER IN BELGIUM* ($\mu g/m^3$)

Study populations		Pb-Air
1 Survey (1974)	<1 km	4.06
	2.5 km	1.00
	Rural	0.29
2 Survey (1975)	<1 km	2.94
	2.5 km	0.74
	Rural	
3 Survey (1976)	<1 km	3.67
	2.5 km	0.80
	Urban	0.45
	Rural	0.30
4 Survey (1977)	<1 km	3.42
	2.5 km	0.49
5 Survey (1978)	<1 km	2.68
	2.5 km	0.54
	Urban	0.56
	Rural	0.37

*Additional airborne data in rural and urban areas obtained as controls are also shown.

Source: Roels et al. (1980).

such as nail-biting and thumb-sucking, as well as eating with the hands, may be an important exposure pathway. It was concluded that intake from contaminated hands contributes at least two to four times as much lead as inhalation of airborne material.

7C.1.5 Meza River Valley, Yugoslavia

In 1967, work was initiated in the community of Zerjav, situated in the Slovenian Alps on the Meza River, to investigate contamination by lead of the air, water, snow, soil, vegetation, and animal life, as well as the human population. The smelter in this community produces about 20,000 metric tons of lead annually; until 1969 the stack emitted lead oxides without control by filters or other devices. Five sampling sites with high-volume samplers operating on a 24-hr basis were established in the four principal settlements within the Meza River Valley (Figure 7C-2): (1) Zerjav, in the center, the site of the smelter, housing 1503 inhabitants, (2) Rudarjevo, about 2 km to the south of Zerjav with a population of 100; (3) Crna, some 5 km to the southwest, population 2198, where there are two sites (Crna-SE and Crna-W); and (4) Mezica, a village about 10 km to the northwest of the smelter with 2515 inhabitants. The data in Table 7C-3 are sufficient to depict general environmental contamination of striking proportions.

7C.1.6 Kosova Province, Yugoslavia

Popovac et al. (1982) discuss lead exposure in an industrialized region near the town of Kosova Mitrovica, Yugoslavia, containing a lead smelter and refinery, and a battery factory. In 1979, 5800 kg of lead were emitted daily from the lead smelter alone. Ambient air concentrations in the town were in the range 21.2-29.2 μg/m^3 in 1980, with levels occasionally reaching 70 μg/m^3. The authors report elevated blood lead levels in most of the children tested; some extremely high values were found, suggesting the presence of congenital lead poisoning.

7C.1.7 Czechoslovakia

Wagner et al. (1981) measured total suspended particulate and airborne lead concentrations in the vicinity of a waste lead processing plant in Czechoslovakia. Data are shown in Table 7C-4. Blood lead levels in 90 children living near the plant were significantly greater than in 61 control children.

7C.1.8 Australia

Heyworth et al. (1981) examined child response to lead in the vicinity of a lead sulfide mine in Northhampton, Western Australia. Two samples of mine tailings measured in 1969 contained 12,000 μg/g and 28,000 μg/g lead; several additional samples analyzed in 1978 contained 22,000-157,000 μg/g lead. Surface soil from the town boundary contained 300 μg/g, while a playground and a recreational area had soil containing 11,000 μg/g and 12,000 μg/g lead, respectively.

Blood lead levels measured in Northhampton children, near the mine, were slightly greater than levels measured in children living a short distance away. The Northhampton blood lead levels were also slightly greater than those reported for children in Victoria, Australia (DeSilva and Donnan, 1980). Heyworth et al. (1981) concluded that the mine tailings could have increased the lead exposure of children living in the area.

7C.2 BATTERY FACTORIES

7C.2.1 Southern Vermont

Watson et al. (1978) investigated homes of employees of a lead storage battery plant in southern Vermont in August and September, 1976. Lead levels in household dust, drinking

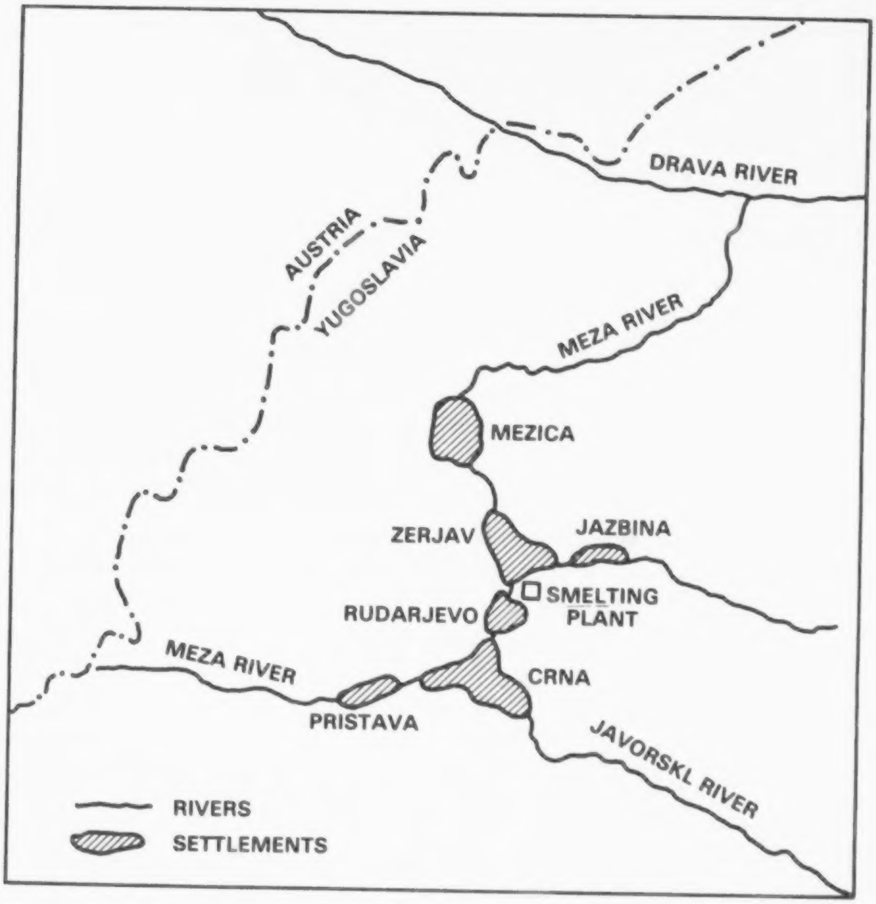
DRAVA RIVER
AUSTRIA
YUGOSLAVIA
MEZA RIVER
MEZICA
ZERJAV
JAZBINA
SMELTING PLANT
RUDARJEVO
MEZA RIVER
CRNA
PRISTAVA
JAVORSKL RIVER
RIVERS
SETTLEMENTS

Figure 7C-2. Schematic plan of lead mine and smelter from Meza Valley, Yugoslavia, study.

Source: Fugas (1977).

TABLE 7C-3. ATMOSPHERIC LEAD CONCENTRATIONS (24-hour) IN THE MEZA VALLEY, YUGOSLAVIA, NOVEMBER 1971 TO AUGUST 1972
($\mu g/m^3$)

Site	Minimum	Maximum	Average
Mezica	0.1	236.0	24.2
Zerjav	0.3	216.5	29.5
Rudarjevo	0.5	328.0	38.4
Crna SE	0.1	258.5	33.7
Crna W	0.1	222.0	28.4

Source: Fugas (1977).

TABLE 7C-4. CONCENTRATIONS OF TOTAL AIRBORNE DUST AND OF AIRBORNE LEAD IN THE VICINITY OF A WASTE LEAD PROCESSING PLANT IN CZECHOSLOVAKIA, AND IN A CONTROL AREA INFLUENCED PREDOMINANTLY BY AUTOMOBILE EMISSIONS

		TSP	Lead
Exposed	n	300	303
	$\bar{x}$ ($\mu g/m^3$)	113.6	1.33
	S	83.99	1.9
	range	19.7-553.4	0.12-10.9
	95% c.i.	123.1-104.1	1.54-1.11
Control	n	56.0	87
	$\bar{x}$ ($\mu g/m^3$)	92.0	0.16
	S	40.5	0.07
	range	10-210	0.03-0.36
	95% c.i.	102.7-81.3	0.17-0.14

n = number of samples; $\bar{x}$ = mean of 24-hour samples;

s = standard deviation; 95% confidence interval.

Source: Wagner et al. (1981).

water, and paint were determined for 22 workers' homes and 22 control homes. The mean lead concentration in dust in the workers' homes was 2,200 µg/g, compared with 720 µg/g in the control homes. Blood lead levels in the workers' children were greater than levels in the control children, and were significantly correlated with dust lead concentrations. No significant correlations were found between drinking water lead and blood lead, or between paint lead and blood lead. It is noteworthy that although 90 percent of the employees showered and changed clothes at the plant, 87 percent brought their work clothes home for laundering. The authors concluded that dust carried home by the workers contributed to increased lead absorption in their children.

7C.2.2 North Carolina

Several cases of elevated environmental lead levels near point sources in North Carolina have been reported by Dolcourt et al. (1978, 1981). In the first instance, dust lead was measured in the homes of mothers employed in a battery factory in Raleigh; blood lead levels in the mothers and their chldren were also measured. Carpet dust was found to contain 1,700-48,000 µg/g lead in six homes where the children had elevated blood lead levels (>40 µg/dl). The authors concluded that lead carried home on the mothers' clothing resulted in increased exposure to their children (Dolcourt et al., 1978). In this particular plant, no uniforms or garment covers were provided by the factory; work clothing was worn home.

In a second case, discarded automobile battery casings from a small-scale lead recovery operation in rural North Carolina were brought home by a worker and used in the family's wood-burning stove (Dolcourt et al., 1981). Two samples of indoor dust yielded 13,000 and 41,000 µg/g lead. A three-year-old girl living in the house developed encephalopathy resulting in permanent brain damage.

In a third case, also in rural North Carolina, a worker employed in an automobile battery reclamation plant was found to be operating an illicit battery recycling operation in his home. Reclaimed lead was melted on the kitchen stove. Soil samples obtained near the house measured as high as 49 percent lead by weight; the driveway was covered with fragments of battery casings. Although no family member had evidence of lead poisoning, there were unexplained deaths among chickens who fed where the lead waste products were discarded (Dolcourt et al., 1981).

7C.2.3 Oklahoma

Morton et al. (1982) studied lead exposure in children of employees at a battery manufacturing plant in Oklahoma. A total of 34 lead-exposed children and 34 control children were examined during February and March, 1978; 18 children in the lead-exposed group had elevated blood lead levels (>30 µg/dl), while none of the controls were in this category.

It was found that many of the battery factory employees also used lead at home, such as casting lead into fishing sinkers and using leaded ammunition. A significant difference in blood lead levels between the two groups of children was found even when families using lead at home were deleted from the data set. Using the results of personal interviews with the homemaker in each household, the authors concluded that dust carried home by the employees resulted in increased exposure of their children. Merely changing clothes at the plant was deemed insufficient to avoid transporting appreciable amounts of lead home: showering and shampooing, in addition to changing clothes, was necessary.

7C.2.4 Oakland, California

Environmental lead contamination at the former site of a wet-cell battery manufacturing plant in Oakland, California was reported by Wesolowski et al. (1979). The plant was operational from 1924 to 1974, and was demolished in 1976. Soil lead levels at the site measured shortly after demolition are shown in Table 7C-5. The increase in median concentrations with depth suggested that the battery plant, rather than emissions from automobiles, was responsible for the elevated soil lead levels. The levels decreased rapidly below 30 cm depth. The contaminated soil was removed to a sanitary landfill and replaced with clean soil; a park has subsequently been constructed at the site.

TABLE 7C-5. LEAD CONCENTRATIONS IN SOIL AT THE FORMER SITE OF A WET-CELL BATTERY MANUFACTURING PLANT IN OAKLAND, CALIFORNIA
(μg/g)

Depth	N*	Range	Mean	Median
Surface	24	57-96,000	4300	200
15 cm	23	13-4200	370	200
30 cm	24	13-4500	1100	360

*N = number of samples.

Source: Wesolowski et al. (1979).

7C.2.5 Manchester, England

Elwood et al. (1977) measured lead concentrations in air, dust, soil, vegetation, and tap water, as well as in the blood of children and adults, in the vicinity of a large battery factory near Manchester. It was found that lead levels in dust, soil, and vegetation decreased with increasing distance from the factory. Airborne lead concentrations did not show a consistent effect with downwind distance, although higher concentrations were found downwind compared with upwind of the factory. Blood lead levels were greatest in the households of battery factory employees: other factors such as distance from the factory, car ownership, age of house, and presence of lead water pipes were outweighed by the presence of a leadworker in the household. These results strongly suggest that lead dust carried home by the factory employees is a dominant exposure pathway for their families. The authors also discussed the work of Burrows (1976), who demonstrated experimentally that the most important means of lead transport from the factory into the home is via the workers' shoes.

APPENDIX 7D

SUPPLEMENTAL DIETARY INFORMATION FROM THE U.S. FDA TOTAL DIET STUDY

The U.S. Food and Drug Administration published a new Total Diet Food List (Pennington, 1983) based on over 100,000 daily diets from 50,000 participants. Thirty five hundred categories of foods were condensed to 201 adult food categories for 8 age/sex groups. Summaries of these data were used in Section 7.3.1.2 to arrive at lead exposures through food, water, and beverages. For brevity and continuity with the crop data of Section 7.2.2.2.1, it was necessary to condense the 201 categories of the Pennington study to 25 categories in this report.

The preliminary lead concentrations for all 201 items of the food list were provided by U.S. Food and Drug Administration (1985). These data represent four Market Basket Surveys, each from a different geographic location. Means of these values have been calculated by EPA, using one-half the detection limit for values reported below detection limit. These data appear in Table 7D-1.

In condensing the 201 categories of Table 7D-1 to the 9 categories of Table 7-17, combinations and fractional combinations of categories were made according to the scheme of Table 7D-2. In this way, specific categories of food more closely identified with farm products were summarized. The assumptions made concerning the ingredients in the final product, (mainly water, flour, eggs, and milk) had little influence on the outcome of the summarization.

TABLE 7D-1. FOOD LIST AND PRELIMINARY LEAD CONCENTRATIONS

Category	Food	Lead concentration* (μg/g)								Mean
1	Whole milk					T				0.003
2	Low fat milk	0.02	T	T	T					0.007
3	Chocolate milk			0.04		T			0.02	0.010
4	Skim milk					T		0.02	0.02	0.008
5	Buttermilk					T	0.09		0.02	0.016
6	Yogurt, plain					0.03				0.006
7	Milkshake	0.06	0.05		0.03	0.05	0.05	0.05	0.03	0.040
8	Evaporated milk	0.08	0.07	0.18	0.10	0.03	0.07	0.07	0.06	0.083
9	Yogurt, sweetened	0.04				T			T	0.009
10	Cheese, American	0.03				T	0.03		0.05	0.016
11	Cottage cheese	0.05				T	0.02		0.02	0.014
12	Cheese, Cheddar	0.04				0.04			0.08	0.021
13	Beef, ground		0.11							0.016
14	Beef, chuck roast	0.09		0.03					0.02	0.019
15	Beef, round steak								0.04	0.007
16	Beef, sirloin									0.002
17	Pork, ham		0.03							0.006
18	Pork chop		0.03							0.006
19	Pork sausage	0.03	0.05				0.04		0.04	0.021
20	Pork, bacon	0.05	0.22							0.035
21	Pork roast								0.03	0.006
22	Lamb chop		0.03							0.006
23	Veal cutlet							0.02	0.04	0.009
24	Chicken, fried	0.04				0.02			0.02	0.011
25	Chicken, roasted					0.03	0.03		0.03	0.013
26	Turkey, roasted									0.002
27	Beef liver	0.11	0.12		0.08	0.06	0.06	0.14	0.09	0.083
28	Frankfurters									0.002
29	Bologna	0.02				0.04			0.05	0.015
30	Salami							0.03	0.06	0.013
31	Cod/haddock filet		0.07					0.08	0.03	0.024
32	Tuna, canned	0.18	0.27	0.08		0.37	0.24	0.06	0.07	0.159
33	Shrimp			0.10		0.05		0.04	0.04	0.030
34	Fish sticks, frozen		0.03					0.02	0.02	0.010
35	Eggs, scrambled									0.002
36	Eggs, fried	0.03							0.07	0.014
37	Eggs, soft boiled						0.06		0.03	0.013

TABLE 7D-1. (continued)

Category	Food	Lead concentration* (μg/g)								Mean
38	Pinto beans, dried	0.04	0.02				T			0.010
39	Pork and beans, canned	0.41	0.07	0.04	0.18	0.06	0.06		0.22	0.130
40	Cowpeas, dried								0.03	0.006
41	Lima beans, dried		0.03			0.02			0.02	0.010
42	Lima beans, frozen		0.03							0.006
43	Navy beans, dried	0.03								0.006
44	Red beans, dried	0.02	0.06				T			0.013
45	Peas, green, canned	0.14	0.28	0.25	0.20	0.11	0.10			0.136
46	Peas, green, frozen	0.03	0.08					0.12		0.030
47	Peanut butter	0.15							0.04	0.025
48	Peanuts								0.04	0.007
49	Pecans	0.03			0.03				0.08	0.019
50	Rice, white	0.05	0.19							0.032
51	Oatmeal	0.06								0.009
52	Farina	0.03				T				0.007
53	Corn grits									0.002
54	Corn, frozen	T	T				0.02			0.006
55	Corn, canned	0.22	0.56	0.06			0.04			0.111
56	Corn, cream style, canned	0.09	0.06	0.11		0.02		0.19	0.34	0.102
57	Popcorn		0.07	0.08		0.04			0.04	0.030
58	White bread				0.03	T	0.03		0.05	0.016
59	Rolls, white	0.03	0.06	0.02	0.35	0.03	0.08	0.04	0.06	0.084
60	Cornbread				0.02	T				0.005
61	Biscuits	0.04		0.02	0.03			0.03		0.016
62	Whole wheat bread	0.05		0.03	0.07	0.03	0.06	T	0.08	0.042
63	Tortilla	0.02	0.03	0.02	0.05	0.04	0.09	0.04	0.04	0.041
64	Rye bread	0.03		0.02	0.04	0.03			0.06	0.023
65	Muffins				0.03	0.03	0.02			0.011
66	Crackers, saltine			0.03	0.03			T	0.03	0.014
67	Corn chips		0.04		0.07			0.10		0.028
68	Pancakes		0.03		0.05				0.04	0.016
69	Noodles	0.04	0.05		0.04		0.03		0.03	0.025
70	Macaroni		0.02		0.03					0.008
71	Corn flakes		0.04							0.007
72	Presweetened cereal		0.06	0.03	0.03	T	0.02		0.08	0.029
73	Shredded wheat cereal					0.03			0.03	0.009
74	Raisin bran cereal			0.03	0.06	0.07	0.02	0.07	0.03	0.036
75	Crisped rice cereal			0.02						0.004

TABLE 7D-1. (continued)

Category	Food	Lead concentration* (μg/g)								Mean
76	Granola	0.03		0.02	0.03	0.02	0.03		0.03	0.021
77	Oat ring cereal	0.03	0.02	0.04	T	0.03			0.03	0.021
78	Apple, raw	0.04	0.04			T	0.02			0.015
79	Orange, raw		0.03	0.02	T	T	0.12	T	T	0.027
80	Banana, raw								0.03	0.006
81	Watermelon, raw			0.02		T	T	0.02		0.009
82	Peach, canned	0.18	0.23	0.28	0.29	0.44	0.12	0.12	0.12	0.223
83	Peach, raw	0.02	0.04			0.05	T	0.02	0.03	0.022
84	Applesauce, canned	0.21	0.19	0.10	0.05		0.10	0.05	0.05	0.094
85	Pear, raw	0.02	0.03		0.04	T	T		0.02	0.017
86	Strawberries, raw	0.03			T	0.02	0.03	0.02	0.04	0.019
87	Fruit cocktail, canned	0.23	0.24	0.13	0.18	0.41	0.22	0.21	0.15	0.221
88	Grapes, raw		0.02		T	T			T	0.007
89	Cantaloupe, raw	0.03	0.08							0.015
90	Pear, canned	0.24	0.22	0.17	0.10	0.17	0.28	0.09	0.08	0.169
91	Plums, raw	T					T		T	0.005
92	Grapefruit, raw	0.03					T	0.01		0.008
93	Pineapple, canned	0.10	0.08	0.05	0.04	0.04	0.04	0.03		0.093
94	Cherries, raw		0.03							0.006
95	Raisins, dried	0.04		0.04	0.05	0.04	0.04		0.05	0.033
96	Prunes, dried	0.05		0.04	0.03	0.04	0.03	0.07	0.04	0.038
97	Avocado, raw	0.03	0.07			0.02	0.04		0.02	0.023
98	Orange juice, frozen	0.02				T	T		T	0.007
99	Apple juice, canned	0.06	0.09	0.02		0.09	0.11		T	0.048
100	Grapefruit juice, frozen	0.03	0.04			T				0.011
101	Grape juice, canned	0.06	0.11	0.04	0.02	0.03	0.11	0.02	0.03	0.053
102	Pineapple juice, canned	0.08	0.02	0.05	0.02	0.03	0.06	0.02	0.04	0.040
103	Prune juice, bottled	0.02		0.02	0.02	0.03	T	T	T	0.015
104	Orange juice, canned	0.05	0.03	0.02	0.03	0.14		0.06	0.09	0.053
105	Lemonade, frozen	0.04	0.07			0.02		T		0.019
106	Spinach, canned	0.80	1.65	0.12	1.34	0.38	0.49	0.25	0.16	0.649
107	Spinach, frozen	0.05	0.10	0.06	0.06	0.14	0.04	0.02	0.06	0.066
108	Collards, frozen	0.05	0.27	0.04	0.03	0.08	0.02	0.04	0.06	0.074
109	Lettuce, raw				0.02	0.04	T		T	0.011
110	Cabbage, raw	0.03			0.02	0.04			T	0.014
111	Coleslaw	0.13				0.02	T		0.04	0.026
112	Sauerkraut, canned	0.77	0.39	0.12	0.64	0.84	0.46	0.93	0.04	0.524
113	Broccoli, frozen	0.04	0.03		T	0.03			T	0.016

TABLE 7D-1. (continued)

Category	Food	Lead concentration* (μg/g)								Mean
114	Celery, raw				T	0.03			0.03	0.010
115	Asparagus, frozen	0.02			T	0.05	T	0.02	T	0.016
116	Cauliflower, frozen				0.04	T				0.008
117	Tomato, raw	0.03			T	0.03				0.010
118	Tomato juice, canned	0.16	0.04	T	0.04	0.10	0.26		0.06	0.084
119	Tomato sauce, canned	0.26	0.31	0.12	0.02	0.10	0.98	0.12	0.15	0.258
120	Tomatoes, canned	0.19	8.20[1]	0.23	0.22	0.38	0.24	0.12	0.15	0.218
121	Beans, snap green, frozen	0.03		0.02	0.02	T	T	T	0.04	0.018
122	Beans, snap green, canned	0.14	0.23	0.12	0.07	0.18	0.05			0.099
123	Cucumber, raw		T		T	0.03			0.04	0.012
124	Squash, summer, frozen	0.04	0.02		0.03	0.02	0.02	T	T	0.019
125	Pepper, green, raw	0.07	0.02		0.02	0.02			0.02	0.020
126	Squash, winter, frozen	0.02			0.02	0.02	T	T	T	0.012
127	Carrots, raw		0.03		0.02	0.02	T	0.02		0.013
128	Onion, raw		0.05	0.02	T	0.04	T	0.01	T	0.019
129	Vegetables, mixed, canned		0.17	0.06	0.05		0.36			0.081
130	Mushrooms, canned	0.25	0.25	0.12	0.37	0.27	0.42	0.25	0.11	0.255
131	Beets, canned	0.17	0.11	0.08	0.12	0.10	T	0.15	0.08	0.103
132	Radish, raw	0.03	0.03		T		0.02		T	0.013
133	Onion rings, frozen	0.07	0.02		T	0.04			0.03	0.022
134	French fries, frozen		T		0.02		T			0.006
135	Mashed potatoes, instant	0.11			0.04					0.020
136	Boiled potatoes, w/o peel		0.02		T					0.005
137	Baked potato, w/ peel		0.04	0.02	0.04		T	0.07		0.023
138	Potato chips	0.03			0.03					0.009
139	Scalloped potatoes	0.04	0.02			0.02	0.02			0.014
140	Sweet potato, baked		0.05	0.04	0.03	0.06	T	0.03	0.03	0.032
141	Sweet potato, candied	0.04	0.04	0.02	0.02	0.05	T	0.02		0.025
142	Spaghetti, w/ meat sauce	0.11	0.12	0.08	0.06	0.04	0.17	0.12	0.39	0.136
143	Beef and vegetable stew		T				T	T		0.005
144	Pizza, frozen	0.06	0.03			0.02	0.02	T	0.02	0.021
145	Chili, beef and beans	0.12	0.05		0.11	0.02	T	0.04	0.46	0.102
146	Macaroni and cheese					0.02				0.004
147	Hamburger sandwich	0.02			0.03	0.07				0.016
148	Meatloaf	0.06	0.46			0.04	0.08	0.06	0.04	0.093

[1] This finding was not included in the calculation of the mean, since it is completely atypical of the lead levels that have been found in canned tomatoes in recent years.

TABLE 7D-1. (continued)

Category	Food	Lead concentration* (μg/g)								Mean
149	Spaghetti in tomato sauce, canned	0.06	0.02			0.03	T			0.016
150	Chicken noodle casserole		0.04			0.03		0.02	0.04	0.017
151	Lasagne	0.11	0.06	0.03	0.06	0.03	0.10	0.10	0.07	0.070
152	Potpie, frozen	0.04	0.03				T		T	0.012
153	Pork chow mein	0.32	0.03	0.04	0.04	0.04	0.04	0.05	0.05	0.076
154	Frozen dinner							0.03	0.04	0.010
155	Chicken noodle soup, canned	0.02	0.02	0.06	0.02	0.04	0.10	0.04	0.05	0.044
156	Tomato soup, canned	0.07	0.02	T	T	0.02	0.03	0.08		0.030
157	Vegetable beef soup, canned	0.04	0.04	0.04	0.07	0.08	0.07	0.06	0.18	0.073
158	Beef bouillon, canned		0.02		0.03	0.05	0.04	T	T	0.021
159	Gravy mix	0.02			T					0.005
160	White sauce	0.05	0.02						0.03	0.014
161	Pickles	0.10	0.09		T	0.04	0.02	T	0.08	0.044
162	Margarine	0.06	0.06							0.017
163	Salad dressing	0.03	0.06							0.013
164	Butter		0.14							0.019
165	Vegetable oil									0.002
166	Mayonnaise				0.06				0.15	0.028
167	Cream	0.06			T					0.010
168	Cream substitute	0.10	0.04						0.04	0.024
169	Sugar	0.07	0.05							0.017
170	Syrup	0.06								0.009
171	Jelly		0.05							0.008
172	Honey	0.12	0.06				0.06			0.031
173	Catsup			0.02	T		0.02	0.02		0.010
174	Ice cream	0.03	0.02	0.03	0.02	0.07	T	0.02	0.15	0.044
175	Pudding, instant					0.04	T			0.008
176	Ice cream sandwich	0.05	0.02			0.02	0.08	0.29		0.058
177	Ice milk	0.07	0.04	0.02		0.02	0.02	T		0.023
178	Chocolate cake	0.13	0.03		0.03	0.06		0.02		0.035
179	Yellow cake	0.16				0.03				0.025
180	Coffee cake	0.04	0.03	0.05	0.05	0.03	0.03		0.09	0.040
181	Doughnuts	0.02			0.03			0.02		0.010
182	Danish pastry	0.06			0.04	0.04	0.02	0.03	0.03	0.028
183	Cookies, choc. chip	0.04	0.03	0.03	0.05	0.04	0.03	0.04	0.02	0.035
184	Cookies, sandwich type	0.03	0.03	0.04	0.03	0.07	0.03	0.12	0.03	0.048
185	Apple pie, frozen	0.04		0.02	0.03	0.03		T		0.017

TABLE 7D-1. (continued)

Category	Food	Lead concentration* (μg/g)								Mean
186	Pumpkin pie	0.05	0.02	0.03	0.05	0.03	0.05	0.03	0.06	0.040
187	Candy, milk chocolate	0.09	0.04	0.09	0.11	0.08	0.05	0.05	0.07	0.073
188	Candy, caramels		0.04	0.04	0.05	0.02	0.06	0.03	0.03	0.034
189	Chocolate powder	0.06	0.03	0.08		0.07	0.07	0.07	0.06	0.055
190	Gelatin dessert	0.02		T	T					0.006
191	Soda pop, cola, canned		0.02							0.004
192	Soda pop lemon-lime, canned	0.13	0.02	0.02						0.023
193	Soft drink powder		0.02				0.02			0.007
194	Soda pop, cola, low cal., canned	0.05	0.02				T			0.011
195	Coffee, instant									0.002
196	Coffee, instant, decaf.		0.02							0.004
197	Tea									0.002
198	Beer, canned	0.02	0.02							0.007
199	Wine	0.03	0.03	0.03	0.03	0.04	0.03	0.05	0.09	0.041
200	Whiskey	0.02				T				0.005
201	Water	T			0.01					0.004

*Individual values for four Market Basket Surveys. "T" means only a trace detected, missing value means below detection limit.

+Means determined by EPA using 0.002 (½ of detection limit) for values below detection limit and 0.01 for detection of trace value.

TABLE 7D-2. SCHEME FOR THE CONDENSATION OF 201 CATEGORIES OF FOOD FROM TABLE 7D-1 INTO 9 CATEGORIES

	Whole Category	Partial Category
Dairy	1-12, 164, 167, 174, 176, 177	0.1 (68-70, 152), 0.2 (151, 178-187) 0.3 (144, 146), 0.5 (156)
Meat	13-37	0.1 (143, 155), 0.2 (144, 146, 151, 178-187), 0.3 (68-70, 145, 153, 154, 158), 0.4 (152) 0.5 (150), 0.6 (142, 147, 148, 149)
Food Crops	38, 40-44, 46-54, 57-67, 71-81, 83, 85, 86, 88, 89, 91, 92, 94-97, 107-111, 113-117, 121, 123-128, 132-141, 159-163 165, 166, 168-173, 175, 188-190	0.2 (148), 0.3 (142, 144, 146, 149-151), 0.4 (147), 0.5 (143, 152), 0.6 (68-70, 178-187), 0.7 (153, 154)
Canned Foods	39, 45, 55, 56, 82, 84, 87, 90, 93, 106, 112, 118-120, 122, 129-131	0.1 (142, 145, 149), 0.2 (144, 148, 150, 151), 0.5 (155-157)
Canned Juices	99, 101, 102, 104	
Frozen Juices	98, 100, 103, 105	
Soda	191, 192, 194	
Canned Beer	198	
Water	193, 195-197, 199-201	0.1 (151), 0.2 (146), 0.4 (143, 155), 0.5 (157), 0.6 (145), 0.7 (158)

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8. EFFECTS OF LEAD ON ECOSYSTEMS

8.1 INTRODUCTION

8.1.1 Scope of Chapter 8

This chapter describes the potential effects of atmospheric lead inputs on several types of ecosystems. An effect is any condition attributable to lead that causes an abnormal physiological response in individual organisms or that perturbs the normal processes of an ecosystem. A distinction is made among natural, cultivated, and urban ecosystems, and extended discussions are included on the mobility and bioavailability of lead in ecosystems.

There are many reports on the effects of lead on individual populations of plants and animals and a few studies on the effects of lead in simulated ecosystems or microcosms. However, the most realistic studies are those that examine the effects of lead on entire ecosystems, as they incorporate all of the ecological interactions among the various populations and all of the chemical and biochemical processes relating to lead (National Academy of Sciences, 1981). Unfortunately, these studies have also had to cope with the inherent variability of natural systems and the confounding frustrations of large-scale projects. Consequently, there are only a handful of ecosystem studies on which to base this report.

Effects at the ecosystem level are usually seen as a form of stress. In nearly every case of stress caused by pollutants, the initial effect is to cause cytological or biochemical changes in specific cells of individual organisms. McLaughlin (1985) has summarized some of the effects on forest ecosystems that have been caused by air pollutants. Examples of cytological or biochemical changes are reduction in enzyme activity, a change in membrane permeability or osmotic potential, or a loss of organelle integrity. These cellular changes cause some disruption of physiological function, such as photosynthesis, respiration, transpiration, root uptake, the opening and closing of stomata, or a disruption of resource allocation, such as growth, reproduction, or defense mechanisms. In turn, the growth of the individual may be directly or indirectly affected, either in amount, timing, or quality. These effects on the individual can cause a change in the productivity of the entire community. Some of the expected effects on the community as a whole or populations within the community might be reduced growth, increased mortality, unbalanced competition, delayed succession, or reduced rate of reproduction.

Because of the complexity of processes that can affect an ecosystem, it is difficult to predict the mechanism by which a specific air pollutant might influence an ecosystem. General categories of effects are those that predispose an ecosystem to stress, those that incite stress, and those that contribute directly to stress (from Manion, 1981, as modified by McLaughlin, 1985). Examples of predisposition are chronic weakening caused by changes in climate, soil moisture, soil nutrients, or competition. Inciting factors are triggering episodes, such as insect defoliation, frost, drought, mechanical injury or increased salinity. Those factors that directly contribute to effects generally do so by accelerating the processes already taking place, such as infections of bark beetles, canker fungi, viruses, root decay fungi, or increased competition. As a general rule, air pollutants are either predisposing or inciting types of agents and are noticed only when a change is triggered by the effect of the pollutant. The effects of air pollutants may go unnoticed for decades, causing only a chronic weakening that cannot be detected by normal methods of evaluating ecosystem stability.

The principle sources of lead entering an ecosystem include the following: the atmosphere (largely from automotive emissions), paint chips, spent ammunition, the application of fertilizers and pesticides, and the careless disposal of lead-acid batteries or other industrial products. Atmospheric lead is deposited on the surfaces of vegetation as well as on ground and water surfaces. In terrestrial ecosystems, this lead is transferred to the upper layers of the soil surface, where it may be retained for a period of several years. The movement of lead within ecosystems is influenced by the chemical and physical properties of lead and by the biogeochemical processes within the ecosystem. Lead is persistent, but in the appropriate chemical environment, may undergo transformations that affect its solubility (e.g., formation of lead sulfate in soils), its bioavailability (e.g., chelation with humic substances), or its toxicity (e.g., chemical methylation).

Because the effects of lead on ecosystems begin with some initial effect on specific cells of individuals within the ecosystem, there are a number of mechanisms or strategies whereby individuals or populations may have developed a resistance to lead toxicity. Wood (1984) has described six potential strategies for resistance to toxic metals: 1) the cell may pump the metal out through the cell membrane, a process that requires energy; 2) the metal may be enzymatically oxidized or reduced to a less toxic form; 3) the cell may synthesize a polymer to trap and remove the metal; 4) the metal may be bound to the cell surface; 5) the metal may be precipitated as an insoluble metal complex; 6) the metal may be biomethylated and transported through the cell membrane by diffusion, a process that requires less energy than actively pumping. The evidence for the biomethylation of lead is circumstantial at best and clearly not conclusive (Craig and Wood, 1981, Reisinger et al., 1981, Chau, 1986).

The previous Air Quality Criteria for Lead (U.S. Environmental Protection Agency, 1977) recognized the problems of atmospheric lead exposure incurred by all organisms including man. Emphasis in the chapter on ecosystem effects was given to reports of toxic effects on specific groups of organisms, e.g. domestic animals, wildlife, aquatic organisms, and vascular and non-vascular plants. Forage containing lead at 80 µg/g dry weight was reported to be lethal to horses, whereas 300 µg/g dry weight caused lethal clinical symptoms in cattle. This report will attempt to place the data in the context of sublethal effects of lead exposure, to extend the conclusions to a greater variety of domestic animals, and to describe the types and ranges of exposures in ecosystems likely to present a problem for domestic animals.

Research on lead in wildlife has traditionally fallen into the following somewhat artificial categories: waterfowl; birds and small mammals; fish; and invertebrates. In all these categories, no correlation could be made in the 1977 report between toxic effects and environmental concentrations. Some recent toxicity studies have been completed on fish and invertebrates and the data are reported below, but there is still little information on the levels of lead that can cause toxic effects in small mammals or birds.

Information on the relationship between soil lead and plants can be expanded somewhat beyond the 1977 report, primarily due to a better understanding of the role of humic substances in binding lead. Although the situation is extremely complex, it is reasonable to state that most plants cannot survive in soil containing 10,000 μg/g dry weight if the pH is below 4.5 and the organic content is below 5 percent. The specifics of this statement are discussed more extensively in Section 8.3.1.2.

Before 1977, natural levels of lead in environmental media other than soil were not well known. Reports of sublethal effects of lead were sparse and there were few studies of total ecosystem effects. Although several ecosystem studies have been completed since 1977 and many problems have been overcome, it is still difficult to translate observed effects under specific conditions directly to predicted effects in ecosystems. Some of the known effects, which are documented in detail in the appropriate sections, are summarized here.

8.1.1.1 Plants. The basic effect of lead on plants is to stunt growth. This may be through a reduction of photosynthetic rate, inhibition of respiration, cell elongation, or root development, or premature senescence. Lead tolerance in ecotypes suggests some effects on population genetics. All of these effects have been observed in isolated cells or in hydroponically-grown plants in solutions comparable to 1 to 2 μg/g soil moisture. These concentrations are well above those normally found in any ecosystem except near smelters or roadsides. Terrestrial plants take up lead from the soil moisture and most of this lead is retained by the roots. There is some evidence for foliar uptake of lead and little evidence that lead can be translocated freely to the upper portions of the plant. Soil applications of calcium and phosphorus may reduce the uptake of lead by roots.

8.1.1.2 Animals. Lead affects the central nervous system of animals and their ability to synthesize red blood cells. Blood concentrations above 0.4 ppm (40 μg/dl) can cause observable clinical symptoms in domestic animals. Calcium and phosphorus can reduce the intestinal absorption of lead. The physiological effects of lead exposures in laboratory animals are discussed in extensive detail in Chapters 10 and 12 of this document.

8.1.1.3 Microorganisms. There is evidence that lead at environmental concentrations occasionally found near roadsides and smelters [10,000 - 40,000 µg/g dw (dry weight)] can eliminate populations of bacteria and fungi on leaf surfaces and in soil. Many of those microorganisms play key roles in the decomposition food chain. It is likely that the affected microbial populations are replaced by others of the same or different species, perhaps less efficient at decomposing organic matter. There is also evidence that microorganisms can mobilize lead by making it more soluble and more readily taken up by plants. This process occurs when bacteria exude organic acids that lower the pH in the immediate vicinity of the plant root.

8.1.1.4 Ecosystems. There are three known conditions under which lead may perturb ecosystem processes. At soil concentrations of 1,000 µg/g or higher, delayed decomposition may result from the elimination of a single population of decomposer microorganisms. Secondly, at concentrations of 500 - 1,000 µg/g, populations of plants, microorganisms, and invertebrates may shift toward lead-tolerant populations of the same or different species. Finally, the normal biogeochemical process that purifies and repurifies the calcium pool in grazing and decomposer food chains may be circumvented by the addition of lead to vegetation and animal surfaces. This third effect can be measured at all ambient atmospheric concentrations of lead.

Some additional effects may occur due to the uneven distribution of lead in ecosystems. It is known that lead accumulates in soil, especially soil with high organic content. Although no firm documentation exists, it is reasonable to assume the following from the known chemistry of lead in soil: 1) other metals may be displaced from the binding sites on the organic matter; 2) the chemical breakdown of inorganic soil fragments may be retarded by the interference of lead with the action of fulvic acid on iron-bearing crystals; and 3) lead in soil may be in equilibrium with moisture films surrounding soil particles and thus be available for uptake by plants.

To aid the reader in understanding the effects of lead on ecosystems, sections have been included that discuss such important matters as how ecosystems are organized, what processes regulate metal cycles, what criteria are valid in interpreting ecosystem effects, and how soil systems function to regulate the controlled release of nutrients to plants. The informed reader may wish to turn directly to Section 8.3, where the discussion of the effects of lead on organisms begins.

8.1.2 Ecosystem Functions

8.1.2.1 Types of Ecosystems. Based on ambient concentrations of atmospheric lead and the distribution of lead in the soil profile, it is useful to distinguish among three types of ecosystems: natural, cultivated, and urban. Natural ecosystems include aquatic and terrestrial ecosystems that are otherwise unperturbed by man, and those managed ecosystems, such as commercial forests, grazing areas, and abandoned fields, where the soil profile has remained undisturbed for several decades. Cultivated ecosystems include those where the soil profile is frequently disturbed and those where chemical fertilizers, weed killers, and pest-control agents may be added. In urban ecosystems, a significant part of the exposed surface includes rooftops, roadways, and parking lots from which runoff, if not channeled into municipal waste processing plants, is spread over relatively small areas of soil surface. The ambient air concentration of lead in urban ecosystems is 5 - 10 times higher than in natural or cultivated ecosystems (See Chapter 7). Urban ecosystems may also be exposed to lead from other than atmospheric sources, such as paint, discarded batteries, and used motor oil. The effects of atmospheric lead depend on the type of ecosystems examined.

8.1.2.2 Energy Flow and Biogeochemical Cycles. To function properly, ecosystems require an adequate supply of energy, which continually flows through the system, and an adequate supply of nutrients, which for the most part, cycle within the ecosystem. There is evidence that lead can interfere with both of these processes. Energy usually enters the ecosystem in the form of sunlight and leaves as heat of respiration. Stored chemical energy may be transported into or out of an ecosystem (e.g., leaf detritus in a stream) or be retained by the ecosystem for long periods of time (e.g., tree trunks). Energy flow through an ecosystem may give structure to the ecosystem by establishing food webs that efficiently regulate the transfer of energy. Segments of these food webs are called food chains. Energy that flows along a grazing food chain is diverted at each step to the detrital food chain.

Unlike energy, nutrient and non-nutrient elements are recycled by the ecosystem and transferred from reservoir to reservoir in a pattern usually referred to as a biogeochemical cycle (Brewer, 1979, p. 139). The reservoirs correspond approximately to the food webs of energy flow. Although elements may enter (e.g., weathering of soil) or leave the ecosystem (e.g., stream runoff), the greater fraction of the available mass of the element is usually cycled within the ecosystem.

Two important characteristics of a reservoir are the amount of the element that may be stored in the reservoir and the rate at which the element enters or leaves the reservoir. Some reservoirs may contain a disproportionately large amount of a given element. For example, most of the carbon in a forest is bound in the trunks and roots of trees, whereas most of the calcium may be found in the soil (Smith, 1980, p. 316). Some large storage reservoirs, such as soil, are not actively involved in the rapid exchange of the nutrient element, but serve as a reserve source of the element through the slow exchange with a more active reservoir, such as soil moisture. When inputs exceed outputs, the size of the reservoir increases. Increases of a single element may reflect instability of the ecosystem. If several elements increase simultaneously, this expansion may reflect stable growth of the community.

Reservoirs are connected by pathways that represent real ecosystem processes. Figure 8-1 depicts the biogeochemical reservoirs and pathways of a typical terrestrial ecosystem. Most elements, especially those with no gaseous phase, do not undergo changes in oxidation state and are equally available for exchange between any two reservoirs, provided a pathway exists between the two reservoirs. The chemical environment of the reservoir may, however, regulate the availability of an element by controlling solubility or binding strengths. This condition is especially true for soils.

Ecosystems have boundaries. These boundaries may be as distinct as the border of a pond or as arbitrary as an imaginary circle drawn on a map. Many trace metal studies are conducted in watersheds where some of the boundaries are determined by topography. For atmospheric inputs to terrestrial ecosystems, the boundary is usually defined as the surface of vegetation, exposed rock, or soil. The water surface suffices for aquatic ecosystems.

Non-nutrient elements differ little from nutrient elements in their biogeochemical cycles. Quite often, the cycling patterns are similar to those of a major nutrient. In the case of lead, the reservoirs and pathways are very similar to those of calcium.

There are three important questions concerning the effect of lead on ecosystems: Does atmospheric lead interfere with the normal mechanisms of nutrient cycles? How does atmospheric lead influence the normal lead cycle in an ecosystem? Can atmospheric lead interfere with the normal flow of energy through an ecosystem?

8.1.2.3 Biogeochemistry of Lead. Naturally occurring lead from the earth's crust is commonly found in soils and the atmosphere. Lead may enter an ecosystem by weathering of parent rock or by deposition of atmospheric particles. This lead becomes a part of the nutrient medium of plants and the diet of animals. All ecosystems receive lead from the atmosphere. More than 99 percent of the current atmospheric lead deposition is now due to human activities (National Academy of Sciences, 1980). In addition, lead shot from ammunition may be found in many waterways and popular hunting regions, leaded paint chips often occur in older urban regions, and lead in fertilizer may contaminate the soil in agricultural regions.

In prehistoric times, the contribution of lead from weathering of soil was probably about 4 g Pb/ha·yr and from atmospheric deposition about 0.02 g Pb/ha·yr, based on estimates of natural and anthropogenic emissions in Chapter 5 and deposition rates discussed in Chapter 6. Weathering rates are presumed to have remained the same, but atmospheric inputs are believed to have increased to 180 g/ha·yr in natural and some cultivated ecosystems, and 3,000 g/ha·yr in urban ecosystems and along roadways (see Chapter 6). In every terrestrial ecosystem of the Northern Hemisphere, atmospheric lead deposition now exceeds weathering by a factor of at least 10, sometimes by as much as 1,000.

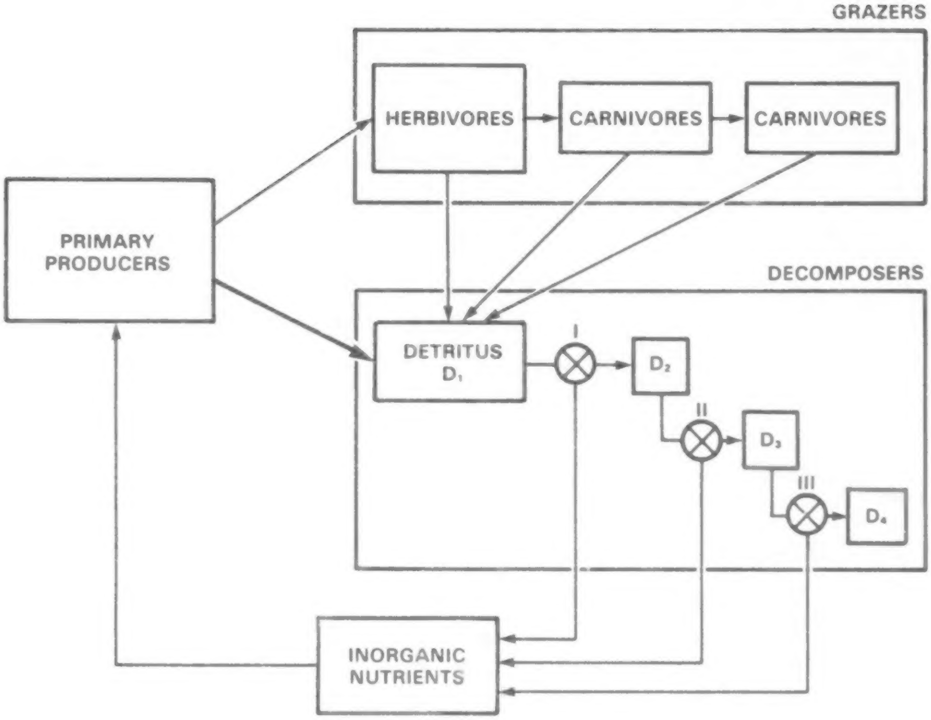
GRAZERS
HERBIVORES
CARNIVORES
CARNIVORES
PRIMARY PRODUCERS
DECOMPOSERS
DETRITUS D₁
I
D₂
II
D₃
III
D₄
INORGANIC NUTRIENTS

Figure 8-1. This figure depicts cycling processes within the major components of a terrestrial ecosystem, i.e. primary producers, grazers and decomposers. Nutrient and non-nutrient elements are stored in reservoirs within these components. Processes that take place within reservoirs regulate the flow of elements between reservoirs along established pathways. The rate of flow is in part a function of the concentration in the preceding reservoir. Lead accumulates in decomposer reservoirs (D_1-D_4) which have a high binding capacity for this metal. When the flow of nutrients is reduced at I, II, or III, the rate of flow of inorganic nutrients to primary producers is reduced.

Source: Adapted from Swift et al. (1979).

Many of the effects of lead on plants, microorganisms, and ecosystems arise from the fact that lead from atmospheric and weathering inputs is retained by soil. Geochemical studies show that less than 3 percent of the inputs to a watershed leave by stream runoff (Siccama and Smith, 1978; Shirahata et al., 1980). In prehistoric times, stream output nearly equalled weathering inputs and the lead content of soil probably remained stable, accumulating at an annual rate of less than 0.1 percent of the original natural lead (reviewed by Nriagu, 1978). Due to human activity, lead in natural soils now accumulates on the surface at an annual rate of 5 - 10 percent of the natural lead. One effect of cultivation is that atmospheric lead is mixed to a greater depth than the 0 - 3 cm of natural soils.

Most of the effects on grazing vertebrates stem from the deposition of atmospheric particles on vegetation surfaces. Atmospheric deposition may occur by either of two mechanisms. Wet deposition (precipitation scavenging through rainout or washout) generally transfers lead directly to the soil. Dry deposition transfers particles to all exposed surfaces. Large particles (>4 μm) are transferred by gravitational mechanisms; small particles (<0.5 μm) are also deposited by wind-related mechanisms.

About half of the foliar dry deposition remains on leaf surfaces following normal rainfall (Elias et al., 1976; Peterson, 1978), but heavy rainfall may transfer the lead to other portions of the plant (Elias and Croxdale, 1980). Koeppe (1981) has reviewed the literature and concluded that less than 1 percent of the surface lead can pass directly into the internal leaf tissues of higher plants. The cuticular layer of the leaves may be an effective barrier to aerosol particles and even to metals in solution on the leaf surface (Arvik and Zimdahl, 1974), and passage through the stomata does not appear to account for a significant fraction of the lead inside leaves (Carlson et al., 1976; 1977).

When particles attach to vegetation surfaces, transfer to soil is delayed from a few months to several years. Due to this delay, large amounts of lead are diverted to grazing food chains, bypassing the soil moisture and plant root reservoirs (Elias et al., 1982).

8 Criteria for Evaluating Ecosystem Effects

As it is the purpose of this chapter to describe the levels of atmospheric lead that may produce adverse effects in plants, animals, and ecosystems, it is necessary to establish the criteria for evaluating these effects. The first step is to determine the connection between air concentration and ecosystem exposure. If the air concentration is known, ecosystem inputs from the atmosphere can be predicted over time and under normal conditions. These inputs and those from the weathering of soil determine the concentration of lead in the nutrient media of plants, animals, and microorganisms. It follows that the concentration of lead in the nutrient medium determines the concentration of lead in the organism and this in turn determines the effects of lead on the organism.

The fundamental nutrient medium of a terrestrial ecosystem is the soil moisture film that surrounds organic and inorganic soil particles. This film of water is in equilibrium with other soil components and provides dissolved inorganic nutrients to plants. It is chemically different than ground water or rain water and there is little reliable information on the relationship between lead in soil and lead in soil moisture. Thus, it appears impossible to quantify all the steps by which atmospheric lead is transferred to plants. Until more information is available on lead in soil moisture, another approach may be more productive. This involves determining the degree of contamination of organisms by comparing the present known concentrations with calculated prehistoric concentrations.

Prehistoric concentrations of lead have been calculated for only a few types of organisms. However, the results are so low that any normal variation, even of an order of magnitude, would not seriously influence the calculation of the degree of contamination. The link between lead in the prehistoric atmosphere and in prehistoric organisms may allow us to predict concentrations of lead in organisms based on present or future concentrations of atmospheric lead.

It is reasonable to infer a relationship between degree of contamination and physiological effect. It seems appropriate to assume that natural levels of lead that were safe for organisms in prehistoric times would also be safe today. It is also reasonable that some additional atmospheric lead can be tolerated by all populations of organisms with no ill effects, that some populations are more tolerant than others, and that some individuals within populations are more tolerant of lead effects than others.

For nutrient elements, the concept of tolerance is not new. The Law of Tolerance (illustrated in Figure 8-2) states that any nutrient may be present at concentrations either too low or too high for a given population and that the ecological success of a population is greatest at some optimum concentration of the nutrient (Smith, 1980, p. 35). In a similar manner, the principle applies to non-nutrient elements. Although there is no minimum concentration below which the population cannot survive, there is a concentration above which the success of the population will decline (point of initial response) and a concentration at which the entire population will die (point of absolute toxicity). In this respect, both nutrients and non-nutrients behave in a similar manner at concentrations above some optimum.

Certain variables make the points of initial response and absolute toxicity somewhat imprecise. The point of initial response depends on the type of response investigated. This response may be at the molecular, tissue, or organismic level, with the molecular response occurring at the lowest concentration. Similarly, at the point of absolute toxicity, death may occur instantly at high concentrations or over a prolonged period of time at somewhat

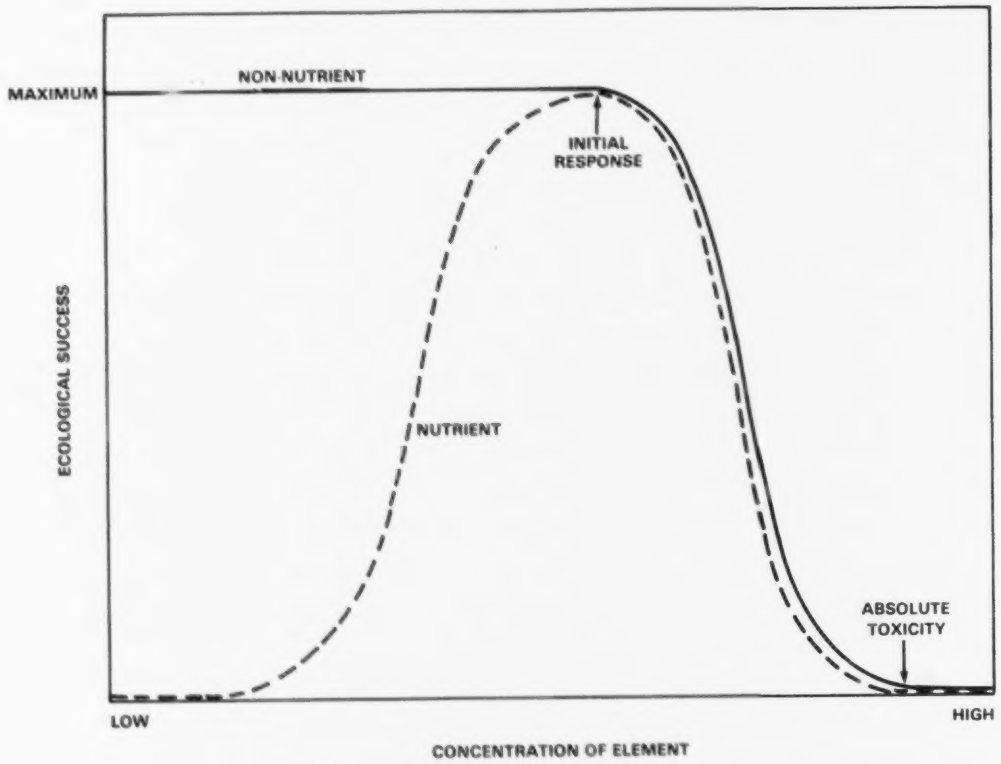

Figure 8-2. The ecological success of a population depends in part on the availability of all nutrients at some optimum concentration. The dashed line of this diagram depicts the rise and decline of ecological success (the ability of a population to grow, survive and reproduce) over a wide concentration range of a single element. The curve need not be symmetrically bell-shaped, but may be skewed to the right or left. Although the range in concentration that permits maximum success may be much wider than shown here, the important point is that at some high concentration, the nutrient element becomes toxic. The tolerance of populations for high concentrations of non-nutrients (solid line) is similar to that of nutrients, although there is not yet any scientific basis for describing the exact shape of this portion of the curve.

Source: Adapted from Smith (1980).

lower concentrations. Nevertheless, the gradient between these two points remains an appropriate basis on which to evaluate known environmental effects, and any information that correctly positions this part of the tolerance curve will be of great value.

The normal parameters of a tolerance curve, i.e., concentration and ecological success, can be replaced by degree of contamination and percent physiological dysfunction, respectively (Figure 8-3). Use of this method of expressing degree of contamination should not imply that natural levels are the only safe levels. It is likely that some degree of contamination can be tolerated with no physiological effect.

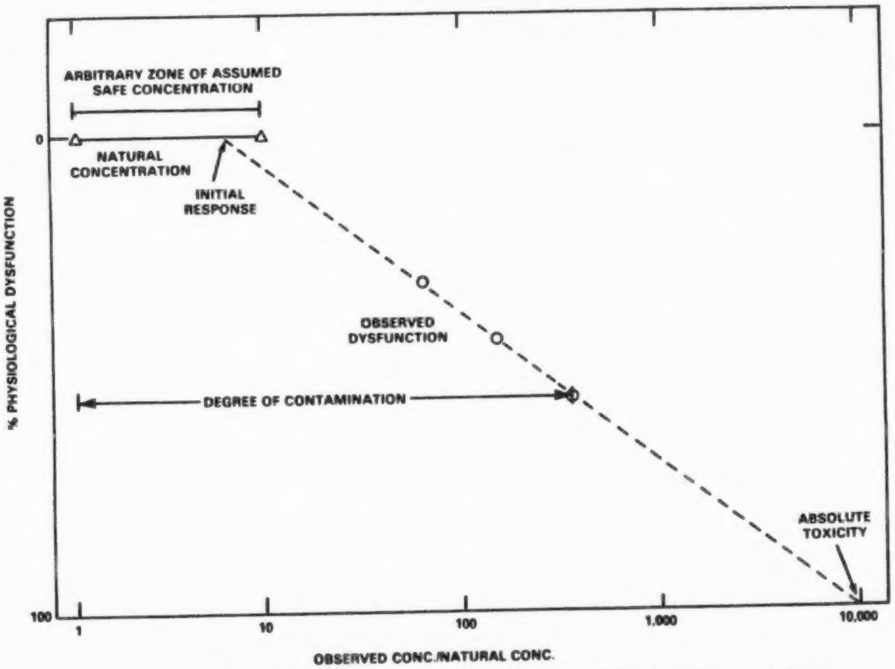

Figure 8-3. This figure attempts to reconstruct the right portion of a tolerance curve, similar to Figure 8-2 but plotted on a semilog scale, for a population using a limited amount of information. If the natural concentration is known for a population and if it is arbitrarily assumed that 10x natural concentration is also safe, then the zone of assumed safe concentration defines the region.

Data reported by the National Academy of Sciences (1980) are used to determine the typical natural lead concentrations shown in various compartments of ecosystems in Table 8-1. These data are from a variety of sources and are simplified to the most probable value within the range reported by NAS. The actual prehistoric air concentration was probably near the low end of the range (0.02-1.0 ng/m^3), as present atmospheric concentrations of 0.3 ng/m^3 in the Southern Hemisphere and 0.07 ng/m^3 at the South Pole (Chapter 5), would seem to preclude natural lead values higher than this.

TABLE 8-1. ESTIMATED NATURAL LEVELS OF LEAD IN ECOSYSTEMS

Component	Range	Best estimate
Air	0.01-1.0 ng/m^3	0.07
Soil		
Inorganic	5-25 µg/g	12.0
Organic	1 µg/g	1.0
Soil moisture	0.0002 µg/g	0.0002
Plant leaves	0.01-0.1 µg/g dw	0.05
Herbivore bones	0.04-0.12 µg/g dw	0.12
Carnivore bones	0.01-0.03 µg/g dw	0.03

Source: Ranges are from the National Academy of Sciences (1980); best estimates are discussed in the text. Units for best estimates are the same as for ranges.

In prehistoric times, the rate of entry of lead into the nutrient pool available to plants was predominantly determined by the rate of weathering of inorganic minerals in fragments of parent rock material. Geochemical estimates of denudation and adsorption rates (Chapter 6) suggest a median value of 12 µg/g as the average natural lead content of total soil, with the concentration in the organic fraction at approximately 1 µg/g.

Studies have shown the lead content of leafy vegetation to be 90 percent anthropogenic, even in remote areas (Crump and Barlow, 1980; Elias et al., 1976, 1978). The natural lead content of nuts and fruits may be somewhat higher than leafy vegetation, based on internal lead concentrations of modern samples (Elias et al., 1982). The natural lead concentrations of herbivore and carnivore bones were reported by Elias et al. (Elias and Patterson, 1980; Elias et al., 1982). These estimates are based on predicted Pb/Ca ratios calculated from the observed biopurification of calcium reservoirs with respect to Sr, Ba, and Pb, on the systematic evaluation of anthropogenic lead inputs to the food chain (Section 8.5.3), and on measurements of prehistoric mammalian bones.

8.2 LEAD IN SOILS AND SEDIMENTS

8.2.1 Distribution of Lead in Soils

Because lead in soil is the source of most effects on plants, microorganisms, and ecosystems, it is important to understand the processes that control the accumulation of lead in soil. The major components of soil are the following: 1) fragments of inorganic parent rock material; 2) secondary inorganic minerals; 3) organic constituents, primarily humic substances, which are residues of decomposition or products of decomposer organisms; 4) Fe-Mn oxide films, which coat the surfaces of all soil particles and appear to have a high binding capacity for metals; 5) soil microorganisms, most commonly bacteria and fungi, although protozoa and soil algae may also be found; and 6) soil moisture, the thin film of water surrounding soil particles that is the nutrient medium of plants. Some watershed studies consider that fragments of inorganic parent rock material lie outside the forest ecosystem, because transfer from this compartment is so slow that much of the material remains inert for centuries.

The concentration of natural lead ranges from 5 to 30 µg/g in the top 5 cm of most soils not adjacent to ore bodies, where natural lead may reach 800 µg/g. Aside from surface deposition of atmospheric particles, plants in North America average about 0.5 - 1 µg/g dw (Peterson, 1978) and animals roughly 2 µg/g (Forbes and Sanderson, 1978). Thus, soils contain the greater part of total ecosystem lead. In soils, lead in parent rock fragments is tightly bound within the crystalline structures of the inorganic soil minerals. It is released to the ecosystem only by surface contact with soil moisture films.

The evidence for atmospheric inputs of lead to soil rests mainly with the accumulation of lead in the soil profile. There are several reports that lead accumulates in the upper layers, usually about 2-5 cm, of the soil, just below the litter layer. This is the soil layer that is usually highest in organic content. Many soils develop by podzolization, characterized by distinct soil horizons caused by the separation and segretation of organic and inorganic compounds, including metal salts and metal-organic complexes. Siccama et al., (1980), and Friedland et al. (1984a, 1984b), found that lead in the forest floors (the litter layer above the mineral soil) of New England have increased during the 1960-1980 at about the same rate that atmospheric concentrations of lead increased. Friedland et al. (1984a, 1984b) found that copper, zinc, and nickel also increased over the same time period, as did the total organic content. They concluded that lead and perhaps other metals may have inhibited decomposition.

Soils adjacent to smelters may be contaminated at a distance of several kilometers away from the source and to a depth of ten or more centimeters. Hogan and Wotton (1984) found elevated concentrations of lead at a distance of 38 kilometers from a Cu-Zn smelter on the surface of the soil, and up to six kilometers at a depth of fifteen centimeters. McNeilly et al. (1984) reported an exponential decrease in lead concentrations of surface soil from 0 to 75 meters for mine spoils. Effects of the spoils were detectable even at a depth of 20 centimeters.

Hutchinson (1980) has reviewed the effects of acid precipitation on the ability of soils to retain cations. Excess calcium and other metals are leached from the A horizon of soils by rain with a pH more acidic than 4.5. Most soils in the eastern United States are normally acidic (pH 3.5 to 5.2) and the leaching process is a part of the complex equilibrium maintained in the soil system. By increasing the leaching rate, acid rain can reduce the availability of nutrient metals to organisms dependent on the top layer of soil. Tyler (1978) reports the effect of acid rain on the leaching rate (reported as residence time) for lead and other metals. Simulated rain of pH 4.2 to 2.8 showed the leaching rate for lead increases with decreasing pH, but not nearly as much as that of other metals, especially Cu, Mn, and Zn. This would be as expected from the high stability constant of lead relative to other metals in humic acids (see Section 6.5.1). It appears from this limited information that acidification of soil may increase the rate of removal of lead from the soil, but not before several major nutrients are removed first. The effect of acid rain on the retention of lead by soil moisture is not known.

8.2.2 Origin and Availability of Lead in Aquatic Sediments

Atmospheric lead may enter aquatic ecosystems by wet or dry deposition (Dolske and Sievering, 1979) or by the erosional transport of soil particles (Baier and Healy, 1977). In waters not polluted by industrial, agricultural, or municipal effluents, the lead concentration is usually less than 1 µg/l. Of this amount, approximately 0.02 µg/l is natural lead and the rest is anthropogenic lead, probably of atmospheric origin (Patterson, 1980). Surface waters mixed with urban effluents may frequently reach lead concentrations of 50 µg/l, and occasionally higher (Bradford, 1977).

In aqueous solution, virtually all lead is divalent, as tetravalent lead can exist only under extremely oxidizing conditions (reviewed by Rickard and Nriagu, 1978; Chapter 3). At pH higher than 5, divalent lead can form a number of hydroxyl complexes, most commonly $PbOH^+$, $Pb(OH)_2$, and $Pb(OH)_3^-$. At pH lower than 5, lead exists in solution as hydrated Pb. In still water, lead is removed from the water column by the settling of lead-containing particulate matter, by the formation of insoluble complexes, or by the adsorption of lead onto suspended organic particles. The rate of sedimentation is determined by temperature, pH, oxidation-reduction potential, ionic competition, the chemical form of lead in water, and certain biological activities (Jenne and Luoma, 1977). McNurney et al. (1977) found 14 µg Pb/g in stream sediments draining cultivated areas and 400 µg/g in sediments associated with urban ecosystems. Small sediment grain size and high organic content contributed to increased retention in sediments.

8.3 EFFECTS OF LEAD ON PLANTS

8.3.1 Effects on Vascular Plants and Algae

Some physiological and biochemical effects of lead on vascular plants have been detected under laboratory conditions at concentrations higher than normally found in the environment. The commonly reported effects are the inhibition of photosynthesis, respiration, or cell elongation, all of which reduce the growth of the plant (Koeppe, 1981). Lead may also induce premature senescence, which may affect the long-term survival of the plant or the ecological success of the plant population. To provide a meaningful evaluation of these effects, it is necessary to examine the correlation between laboratory conditions and typical conditions in nature with respect to form, concentration, and availability of lead. First, the reader must understand what is known of the movement of lead from soil to the root to the stem and finally to the leaf or flower. Most notably, there are specific barriers to lead at the soil:soil moisture interface and at the root:shoot interface that retard the movement of lead and reduce the impact of lead on photosynthetic and meristematic (growth and reproduction) tissue.

8.3.1.1 Uptake by Plants. Most of the lead in or on a plant occurs on the surfaces of leaves and the trunk or stem. The surface concentration of lead in trees, shrubs, and grasses exceeds the internal concentration by a factor of at least five (Elias et al., 1978). Foliar uptake was believed to account for less than 1 percent of the uptake by roots (Arvik and Zimdahl, 1974; reviewed by Koeppe, 1981; Zimdahl, 1976). Krause and Kaiser (1977) were able to show foliar uptake and translocation of lead mixed with cadmium, copper, and manganese oxides when applied in extremely large amounts (122 mg/m^2) directly to leaves. This would be comparable to 100,000 days accumulation at a remote site (0.12 $ng/cm^2 \cdot d$) (Elias et al., 1978). However, recent isotopic evidence by Facchetti and Geiss (1982) and Patterson (1982) and mass balance interpretations from watershed data (Lindberg and Harriss, 1981) suggest that lead can be absorbed across the leaf surface into internal plant tissues. Nevertheless, the major effect of surface lead at ambient concentrations seems to be on subsequent components of the grazing food chain (Section 8.4.1) and on the decomposer food chain following litterfall (Elias et al., 1982). (See also Section 8.4.2.)

In the soil, the availability of metals to plants is generally controlled by the concentration and form of the metal, which are in turn influenced by such soil forming processes as gleying, leaching, podzolization, and the accumulation of organic matter at the surface. Other factors such as pH and the presence of other cations may also be important. The amount of lead that enters plants by this route is determined by the availability of lead in soil, with apparent variations according to plant species. Soil cation exchange capacity, a major factor, is determined by the relative size of the clay and organic fractions, soil pH, and the amount of Fe-Mn oxide films present (Nriagu, 1978). Of these, organic humus and high soil pH are the dominant factors in immobilizing lead (see Section 6.5.1). Under natural conditions, most of the total lead in soil would be tightly bound within the crystalline structure of inorganic soil fragments, unavailable to soil moisture. Available lead, bound on clays, organic colloids, and Fe-Mn films, would be controlled by the slow release of bound lead from inorganic rock sources. Since before 3000 B.C., atmospheric lead inputs through litter decomposition have increased the pool of available lead bound on organic matter within the soil reservoir (see Section 5.1).

Because lead is strongly immobilized by humic substances, only a small fraction (perhaps 0.01 percent in soils with 20 percent organic matter, pH 5.5) is released to soil moisture (see Section 6.5.1). In soil moisture, lead may pass along the pathway of water and nutrient uptake on either a cellular route through the cell membranes of root hairs (symplastic route) or an extracellular route between epidermal cells into the intercellular spaces of the root cortex (apoplastic route) (Foy et al., 1978). Lead probably passes into the symplast by membrane transport mechanisms similar to the uptake of calcium or other bivalent cations.

In soils with lead concentrations within the range of natural lead (15-30 µg/g), only trace amounts of lead are absorbed by plants. The amount absorbed increases when the concentration of lead in soil increases or when the binding capacity of soil for lead decreases. Uptake by root systems does not necessarily mean the lead reaches the stems, leaves or fruits. Rather, the process should be seen as a soil-plant continuum that strongly favors retention of lead by the soil and the root system.

When viewed from the perspective of the uptake of nutrients such as calcium, there are at least three mechanisms whereby lead can be taken up by roots: transpirational mass flow, diffusion, and active transport (Jenny, 1980). Probably the most significant is transpirational mass flow. In the process of absorbing and transporting water from the soil to the leaves, the plant absorbs relatively large amounts of ions in solution. Since plants take up about 100 times their weight in water each growing season, this process could account for twice the normal amounts of lead found in vegetation, assuming equilibrium between the soil and the soil solution. For example, a lettuce plant transpired 100 liters of water during the season, which contained 5000 µg of lead (Rabinowitz, 1972). But the plant itself contained only 2500 µg, most of which was in the roots.

Diffusion can occur along a concentration gradient whenever the transpiration stream is idle, e.g., during the night or during periods of high humidity. Because the concentration of lead in the soil solution is usually higher than in the plant, and because lead bound on the cellulose matrix of the cell wall would not effect the concentration gradient, the flow of lead would probably be toward the root. Although the third mechanism, active transport, is an important process for nutrient elements, there is no evidence that such a process occurs for lead or any other non-nutrient. This process requires energy, and it is unreasonable that a plant would expend energy to take up non-nutrient elements.

The soil-root continuum is a complex structure that consists of the soil particles, the soil solution, the mutigel or other remnants of root exudates, the epidermal cells with elongated root hairs, and the root cortial cells. The walls of the epidermal cells are a loose matrix of cellulose and hemicellulose fibers. Much of this continuum is of biological origin and contains compounds active in ion exchange, such as hemicelluloses and pectic substances that are heavily endowed with -COOH groups, and proteins that also have charged groups. As a cation moves from the soil particle to the root cortex, whether by mass flow or diffusion, it is continually proximate to root structures with a high binding capacity. Lead is more tightly bound at these sites than other cations, even calcium. Consequently, relatively little lead passes through the roots into the shoot. It appears that most of the soil lead is retained within the root. However, some plants may allow more lead to translocate than others. Rabinowitz (1972) found that for lettuce and wild oats growing in soils with increasing lead concentrations, the lettuce translocated very little soil lead but the wild oats translocated proportionately greater amounts. The author was able to distinguish isotopically between soil and atmospheric lead, and found also that more than half the lead in plants, after water washing, was of atmospheric origin when the plants were grown at 30 meters from a freeway.

At 500 µg Pb/g nutrient solution, lead has been shown to accumulate in the cell walls of germinating Raphinus sativus roots (Lane and Martin, 1982). This concentration is much higher than that found by Wong and Bradshaw (1982) to cause inhibition of germinating root elongation (less than 2.5 µg/g), absence of root growth (5 µg/g), or 55 percent inhibition of seed germination (20-40 µg/g) in the rye grass, Colium perenne. Lane and Martin (1982) also observed lead in cytoplasmic organelles that because of their osmiophillic properties, appeared to have a storage function. It was suggested that the organelles eventually emptied their contents into the tonoplast.

The accumulation of lead in cell walls and cytoplasmic bodies has also been observed in blue-green algae by Jensen et al. (1982), who used X-ray energy dispersive analysis in conjunction with scanning electron microscopy to observe high concentrations of lead and other metals in these single-celled procaryotic organisms. They found the lead concentrated in the third of the four-layered cell wall and in polyphosphate bodies (not organelles, since they are not membrane-bound) which appeared to be a storage site for essential metals. The nutrient solution contained 100 µg Pb/g. The same group (Rachlin et al., 1982) reported morphological changes in the same blue-green alga (Plectonema boryanum). There was a significant increase in cell size caused by the lead, which indicated that the cell was able to detoxify its cytoplasm by excreting lead with innocuous cell wall material.

It appears that two defensive mechanisms may exist in the roots of plants for removing lead from the stream of nutrients flowing to the above-ground portions of plants: lead may be deposited with cell wall material exterior to the individual root cells, or may be sequestered in organelles within the root cells. Any lead not captured by these mechanisms would likely move with nutrient metals cell-to-cell through the symplast and into the vascular system.

Uptake of lead by plants may be enhanced by symbiotic associations between plant roots and mycorrhizal fungi. The three primary factors that control the uptake of nutrients by plants are the surface area of the roots, the ability of the root to absorb particular ions, and the transfer of ions through the soil. The symbiotic relationship between mycorrhizal fungi and the roots of higher plants can increase the uptake of nutrients by enhancing all three of these factors (Voigt, 1969). The typical ectomycorrhiza consists of a mantle or sheath of mycelia that completely surrounds the root. The physical extension of the sheath may increase the volume of the root two to three times (Voigt, 1969). Mycorrhizal roots often show greater affinities for nutrients than do uninfected roots of the same species grown in the same conditions. In many soil systems, where the bulk of the nutrients are bound up in parent rock material, efficient uptake of these nutrients by plants depends on the ability of organisms in the rhizosphere (plant roots, soil fungi, and bacteria) to increase the rates of weathering. Mycorrhizal fungi are known to produce and secrete into their environment many different acidic compounds (e.g., malic and oxalic acids). In addition, mycorrhizal roots have been shown to release more carbon dioxide into the rhizosphere than do non-mycorrhizal roots as a result of their increased rates of respiration. Carbon dioxide readily combines with soil moisture to produce carbonic acid. All of these acids are capable of increasing the weathering rates of soil particles such as clays, and altering the binding capacity of organic material, thereby increasing the amount of nutrients and other cations in the soil solution. Mycorrhizae are known to enhance the uptake of zinc by pine roots (Bowen et al., 1974), and it is likely that lead uptake is similarly increased, by inference to the ability of mycorrhizae to enhance the uptake of calcium by pine roots (Melin and Nilsson, 1955; Melin et al., 1958).

The translocation of lead to aboveground portions of the plant is not clearly understood. Lead may follow the same pathway and be subject to the same controls as a nutrient metal such as calcium. This assumption implies that the plant root has no means of discriminating against lead during the uptake process, and it is not known that any such discrimination mechanism exists. There may be several mechanisms, however, that excrete lead back out of the root or that prevent its translocation to other plant parts. The primary mechanisms may be storage in cell organelles or adsorption on cell walls. The apoplast contains an important supply of plant nutrients, including water. Lead in the apoplast remains external to the cells and cannot pass to vascular tissue without at least passing through the cell membranes of the endodermis. Because this extracellular region is bounded on all sides by cell walls, the surface of which is composed of layers of cellulose strands, the surface area of the apoplast is comparable to a sponge. It is likely that much of the lead in roots is adsorbed to the apoplast surface. Dictyosomes (cytoplasmic organelles that contain cell wall material) may carry lead from inside the cell through the membrane to become a part of the external cell wall (Malone et al., 1974), possibly replacing calcium in calcium pectate. Lead may also be stored and excreted as lead phosphate in dictyosome vesicles (Malone et al., 1974). Nevertheless, some lead does pass into the vascular tissue, along with water and dissolved nutrients, and is carried to physiologically active tissue of the plant.

Evidence that lead in contaminated soils can enter the vascular system of plants and be transported to above-ground parts may be found in the analysis of tree rings. Rolfe (1974) found fourfold increases in both rural and urban trees, comparing 10-year increments of annual rings for the period 1910-20 to annual rings of the period 1963-73. Symeonides (1979) found a twofold increase from 1907-17 to 1967-77 in trees at a high-lead site, with no increase in trees from a low-lead site. Baes and Ragsdale (1981), using only ring porous species, found significant post-1930 increases in Quercus and Carya with high lead exposure, but only in Carya with low-lead exposure. These chronological records confirm that lead can be translocated from roots to the upper portions of the plant and that the amounts translocated are in proportion to the concentrations of lead in soil.

8.3.1.2 Physiological Effects on Plants. Because most of the physiologically active tissue of plants is involved in growth, maintenance, and photosynthesis, it is expected that lead might interfere with one or more of these processes. Indeed, such interferences have been observed under optimal growth conditions in laboratory experiments at lead concentrations greater than those normally found in the field, except near smelters or mines (Koeppe, 1981). It is likely that because these are the physiological processes studied more vigorously than others, more is known of these effects. Studies of lead effects on other plant processes, especially maintenance, flowering, and hormone development, have not been conducted and no conclusion can be reached concerning these processes.

Inhibition of photosynthesis by lead may be by direct interference with the light reaction or the indirect interference with carbohydrate synthesis. With 21 μg Pb/g reaction solution, Miles et al. (1972) demonstrated substantial inhibition of photosystem II near the site of water splitting, a biochemical process believed to require manganese. Homer et al. (1979) found a second effect on photosystem II at slightly higher concentrations of lead. This effect was similar to that of DCMU [3-(3,4-dichlorophenyl)-1,1-dimethylurea], a reagent commonly used to uncouple the photosynthetic electron transport system. Bazzaz and Govindjee (1974) suggested that the mechanism of lead inhibition was a change in the conformation of the thylakoid membranes, separating and isolating pigment systems I and II. Wong and Govindjee (1976) found that lead also interferes with P700 photooxidation and re-reduction, a part of the photosystem I light reaction. Homer et al. (1981) found a lead tolerant population of the grass Phalaris arundinacea had lowered the ratio of chlorophyll a/chlorophyll b, believed to be a compensation for photosystem II inhibition. There was no change in the total amount of chlorophyll, but the mechanism of inhibition was considered different from that of Miles et al. (1972). Hampp and Lendzian (1974) found that lead chloride inhibits the synthesis of chlorophyll b more than that of chlorophyll a at concentrations up to 100 mg Pb/g. Devi Prasad and Devi Prasad (1982) found 10 percent inhibition of pigment production in three species of green algae at 1 μg/g, increasing to 50 percent inhibition at 3 μg/g. Bazzaz et al. (1974, 1975) observed reduced net photosynthesis which may have been caused indirectly by inhibition of carbohydrate synthesis. Without carbohydrates, stomatal guard cells remain flaccid, transpiration ceases, carbon dioxide fixation decreases, and further carbohydrate synthesis is inhibited.

In the quantification of growth inhibition, one can measure either the concentration of lead in the nutrient medium or in the tissue that is growth inhibited. Lead concentrations in the nutrient medium relate directly to the degree of environmental contamination, but the more precise measurement is in the tissue, since there would be a more direct correlation between the lead concentration and the physiological processes inhibited. Burton et al. (1983) determined that when tissue concentrations in the shoots of Sitka-spruce seedlings exceeded about 20 μg Pb/g dw, growth inhibition became significant, and lethal at about 40 μg/g. This narrow range between the onset of inhibition and lethality was attributed to the sequestering of lead in the roots and shoots up to 19 μg/g, above which any additional lead would be more available and extremely toxic. The stunting of plant growth may be by the inhibition of the growth hormone IAA (indole-3-ylacetic acid). Lane et al. (1978) found a 25 percent reduction in elongation at 10 μg/g lead as lead nitrate in the nutrient medium of wheat coleoptiles. This effect could be reversed with the addition of calcium at 18 μg/g. Lead may also interfere with plant growth by reducing respiration or inhibiting cell division. Miller and Koeppe (1971) and Miller et al. (1975) showed succinate oxidation inhibition in isolated mitochondria as well as stimulation of exogenous NADH oxidation with related mitochondrial swelling. Hassett et al. (1976), Koeppe (1977), and Malone et al. (1978) described significant inhibition of lateral root initiation in corn. Inhibition increased with the simultaneous addition of cadmium.

Sung and Yang (1979) found that lead at 1 μg/g can complex with and inactivate ATPase to reduce the production and utilization of ATP in kidney bean (Phaseolus vulgaris) and buckwheat leaves (Fagopyrum esculentum). The lead was added hydroponically at concentrations up to

1,000 µg/g. Kidney bean ATPase showed a continued response from 1 to 1,000 µg/g, but buckwheat leaves showed little further reduction after 10 µg/g. Neither extracted ATP nor chemically added ATP could be used by the treated plants. Lee et al. (1976) found a 50 percent increase in the activity of several enzymes related to the onset of senescence in soybean leaves when lead was added hydroponically at 20 µg/g. These enzymes were acid phosphatase, peroxidase, and alpha-amylase. A build-up of ammonia was observed along with a reduction in nitrate, calcium, and phosphorus. Glutamine synthetase activity was also reduced by 65 percent. Continued increases in effects were observed up to 100 µg/g, including a build-up of soluble protein. Päivöke (1979) also observed a 60 percent increase in acid phosphatase activity during the first 6 days of pea seedling germination (Pisum sativum) at 2 µg/g, under low nutrient conditions. The accumulation of soluble protein was observed and the effect could be reversed with the addition of nutrients, including calcium.

Scarponi and Perucci (1984) reported that lead can interfere with the synthesis of ALA-dehydratase in corn, but does not appear to affect the activity of this enzyme. This enzyme catalyzes the conversion of δ-aminolevulinic acid to porphobilinogen, an intermediate in chlorophyll synthesis. The concentration of lead was above 10,000 µg/l in the nutrient solution.

The interaction of lead with calcium has been shown by several authors, most recently by Garland and Wilkins (1981), who demonstrated that calcium could partially overcome the effects of lead on growth in barley seedlings (Hordeum vulgare). Seedlings that were growth-inhibited at 2 µg Pb/g sol. with no added calcium, grew at about half the control rate with 17 µg Ca/g sol. This relation persisted up to 25 µg Pb/g sol. and 500 µg Ca/g sol.

Chaney and Strickland (1984) measured the effects of lead on the germination on red pine pollen. Following exposure in an aqueous nutrient medium, two parameters were measured: pollen germination and germ tube elongation. Pollen germination was inhibited by greater than 10 percent only at relatively high concentrations of lead, about 1,000 µg/l, but the most significant effect was shown for germ tube elongation, which showed 10 percent inhibition at about 150 µg/l.

These studies of the physiological effects of lead on plants all show some effect at concentrations from 2 to 10 µg/g in the nutrient medium of hydroponically-grown agricultural plants. It is probable that no effects would have been observed at these concentrations had the lead solutions been added to normal soil, where the lead would have been bound by humic substances. There is no firm relationship between soil lead and soil moisture lead, because each soil type has a unique capacity to retain lead and to release that lead to the soil moisture film surrounding the soil particle. Once in soil moisture, lead seems to pass freely to the plant root according to the capacity of the plant root to absorb water and dissolved substances (Koeppe, 1981).

Chapter 6 discusses the many parameters controlling the release of lead from soil to soil moisture, but so few data are available on observed lead concentrations in soil moisture that no model can be formed. It seems reasonable that there may be a direct correlation between lead in hydroponic media and lead in soil moisture. Hydroponic media typically have an excess of essential nutrients, including calcium and phosphorus, so that movement of lead from hydroponic media to plant root would be equal to or slower than movement from soil moisture to plant root. Hughes (1981) adopted the general conclusion that extractable soil lead is typically 10 percent of total soil lead. However, this lead was extracted chemically under laboratory conditions more rigorous than the natural equilibrium between soil and soil moisture. Ten percent should therefore be considered the upper limit, where the ability of soil to retain lead is at a minimum. A lower limit of 0.01 percent is based on the only known report of lead in both soil and soil moisture (16 µg/g soil, 1.4 ng/g soil moisture; Elias et al., 1982). This single value shows neither trends with different soil concentrations nor the soil component (organic or inorganic) that provides the lead to the soil moisture. But the number (0.01 percent) is a conservative estimate of the ability of soil to retain lead, since the conditions (pH, organic content) were optimum for retaining lead. A further complication is that atmospheric lead is retained at the surface (0-2 cm) of the soil profile (Martin and Coughtrey, 1981), whereas most reports of lead in soil pertain to samples from 0 to 10 cm as the "upper" layer of soil. Any plant that absorbs solely from the top few centimeters of soil obviously is exposed to more lead than one with roots penetrating to a depth of 25 cm or more. Agricultural practices that cultivate soil to a depth of 25 cm blend in the upper layers with lower to create a soil with average lead content somewhat above background.

These observations lead to the general conclusion that even under the best of conditions where soil has the highest capacity to retain lead, most plants would experience reduced growth rate (inhibition of photosynthesis, respiration, or cell elongation) in soils of 10,000 µg Pb/g or greater. Khan and Frankland (1983) observed stunted growth in radish plants at 1000 µg Pb/g soil when the lead was added as chloride, with complete growth inhibition at 5000 µg/g. The effects were less severe when lead oxide was added to the soil. Concentrations approaching these values typically occur around smelters (Martin and Coughtrey, 1981) and near major highways (Wheeler and Rolfe, 1979). These conclusions pertain to soil with the ideal composition and pH to retain the maximum amount of lead. Acid soils or soils lacking organic matter would inhibit plants at much lower lead concentrations.

The rate at which atmospheric lead accumulates in soil varies from 1.1 $mg/m^2 \cdot yr$ average global deposition (Table 6-6) to 3,000 $mg/m^2 \cdot yr$ near a smelter (Patterson et al., 1975). Assuming an average density of 1.5 g/cm^3, undisturbed soil to a depth of 2 cm (20,000 cm^3/m^2) would incur an increase in lead concentration at a rate of 0.04 to 100 µg/g soil·yr. This means remote or rural area soils may never reach the 10,000 µg/g threshold but that undisturbed soils closer to major sources may be within range in the next 50 years.

8.3.1.3 *Lead Tolerance in Vascular Plants*. Some plant species have developed populations tolerant to high-lead soils (Antonovics et al., 1971). In addition to Homer et al. (1981) cited above, Jowett (1964) found populations of *Agrostis tenuis* in pure stands on acidic spoil banks near an abandoned mine. The exclusion of other species was attributed to root inhibition. Populations of *A. tenuis* from low-lead soils had no tolerance for the high-lead soils. Several other studies suggest that similar responses may occur in populations growing in lead-rich soils (reviewed in Peterson, 1978). A few have suggested that crops may be cultivated for their resistance to high-lead soils (Gerakis et al., 1980; John, 1977).

Using populations taken from mine waste and uncontaminated control areas, some authors have quantified the degree of tolerance of *Agrostis tenuis* (Karataglis, 1982) and *Festuca rubra* (Wong, 1982) under controlled laboratory conditions. Root elongation was used as the index of tolerance. At 36 µg Pb/g nutrient solution, all populations of *A. tenuis* were completely inhibited. At 12 µg Pb/g, the control populations from low-lead soils were completely inhibited, but the populations from mine soils achieved 30 percent of their normal growth (growth at no lead in nutrient solution). At 6 µg/g, the control populations achieved 10 percent of their normal growth; tolerant populations achieved 42 percent. There were no measurements below 6 µg/g. Wong (1982) measured the index of tolerance at one concentration only, 2.5 µg Pb/g nutrient solution, and found that non-adapted populations of *Festuca rubra* that had grown on soils with 47 µg/g total lead content were completely inhibited, populations from soils with 350 - 650 µg/g achieved 3-7 percent of normal growth, and populations from 5,000 µg/g soil achieved nearly 40 percent of normal growth. Tolerance indices should be used with caution because they depend on two measurements that may be genetically independent. Humphreys and Nicholls (1984) suggested that different genes regulated root elongation in a control solution and in the heavy-metal solution.

These studies support the conclusion that inhibition of plant growth begins at a lead concentration of less than 1 µg/g soil moisture and becomes completely inhibitory at a level between 3 and 10 µg/g. Plant populations that are genetically adapted to high-lead soils may achieve 50 percent of their normal root growth at lead concentrations above 3 µg/g. These experiments did not show the effect of reduced root growth on total productivity, but they did show that exposure to high-lead soils is a requirement for genetic adaptation and that, at least in the case of *F. rubra*, plant lead concentrations increase with increasing concentrations in the soil.

There are a few plants known to be hyperaccumulators of metals (Reeves and Brooks, 1983). These plants appear to show no adverse effects even when their tissue concentrations reach

1000 µg/g dry weight. About 100 species of plants are known to hyperaccumulate nickel, fifteen each for copper and cobalt. Reeves and Brooks (1983) describe two species that hyperaccumulate lead and mention three others reported in the literature. The fact that many of these species belong to the genus Alyssum suggests a genetic mechanism of metal tolerance.

8.3.1.4 Effects of Lead on Forage Crops. In the 1977 Air Quality Criteria Document for Lead (U.S. Environmental Protection Agency, 1977), there was a general awareness that most of the lead in plants was surface lead from the atmosphere. Most studies since then have addressed the problem of distinguishing between surface and internal plant lead. The general conclusion is that, even in farmlands remote from major highways or industrial sources, 90 - 99 percent of the total plant lead is of anthropogenic origin (National Academy of Sciences, 1980). Obviously, the critical agricultural problem concerns forage crops and leafy vegetables. In Great Britain, Crump and Barlow (1982) determined that, within 50 m of the highway, surface deposition is the major source of lead in forage vegetation. Beyond this range, seasonal effects can obscure the relative contribution of atmospheric lead. The atmospheric deposition rate appears to be much greater in the winter than in the summer. Two factors may explain this difference. First, deposition rate is a function of air concentration, particle size distribution, windspeed, and surface roughness. Of these, only particle size distribution is likely to be independent of seasonal effects. Lower windspeeds or air concentration during the summer could account for lower deposition rates. Second, it may be that the deposition rate only appears to change during the summer. With an increase in biomass and a greater turnover in biomass, the effective surface area increases and the rate of deposition, which is a function of surface area, decreases. During the winter, lead may not build up on the surface of leaves as it does in summer, even though the flux per unit of ground area may be the same.

8.3.1.5 Effects on Algae. Sicko-Goad and Lazinsky (1981) have presented cytological evidence that lead can be incorporated into polyphosphate bodies in some algal species (Diatoma tenue var. elongatum, Scenedesmus sp.), presumably as a tolerance mechanism. They also report the immobilization of lead in cell vacuoles. At high concentrations (207 µg/g), Roderer (1984a) found deformations of cell organelles, especially nuclei and mitochondria, and increased autolytic activity in the chrysophyte Poterioochromonas malhamensis, a unicellular alga. In the same study, organolead compounds, TriEL and TEL were found to cause an increase in number and size of nuclei, contractile vacuoles, chloroplasts and dictyosomes, as well as a marked accumulation of lipid droplets and lysosomes. The concentrations for these effects were 10 µM TriEL and 100 µM TEL. Similar results were reported in a review of the toxic effects of organolead compounds by Roderer (1984b).

8.3.1.6 Summary of Plant Effects. When soil conditions allow lead concentrations in soil moisture to exceed 2 - 10 µg/g, most plants experience reduced growth due to the inhibition of one or more physiological processes. Excess calcium or phosphorus may reverse the effect. Plants that absorb nutrients from deeper soil layers may receive less lead. Acid rain is not likely to release more lead until after major nutrients have been depleted from the soil. A few species of plants have the genetic capability to adapt to high lead soils.

8.3.2 Effects on Bacteria and Fungi

Wood and Wang (1983) discuss possible mechanisms for microbial resistance to metals, noting that some metals (e.g., Al, Pb, Sn, Be) occur at crustal abundances greater than other metals known to be required nutrients. Abundance alone is not a sufficient condition for the evolution of a nutritive requirement. A second condition is solubility in anaerobic conditions. Except at low pH, aluminum, lead, and tin are insoluble in an anaerobic solution and would not have been available to primitive microorganisms during the early stages of their evolution.

8.3.2.1 Effects on Decomposers. Tyler (1972) explained three ways in which lead might interfere with the normal decomposition processes in a terrestrial ecosystem. Lead may be toxic to specific groups of decomposers, it may deactivate enzymes excreted by decomposers to break down organic matter, or it may bind with the organic matter to render it resistant to the action of decomposers. Because lead in litter may selectively inhibit decomposition by soil bacteria at 2,000 - 5,000 µg/g (Smith, 1981, p. 160), forest floor nutrient cycling processes may be seriously disturbed near lead smelters (Bisessar, 1982; Watson et al., 1976). This is especially important because approximately 70 percent of plant biomass enters the decomposer food chain (Swift et al., 1979, p. 6). If decomposition of the biomass is inhibited, then much of the energy and nutrients remain unavailable to subsequent components of the food chain. There is also the possibility that the ability of soil to retain lead would be reduced, as humic substances are byproducts of bacterial decomposition.

Babich et al. (1983) introduced the concept of ecological dose as it applies to the effects of metals on ecological processes in soil. The inhibition of microbe-mediated processes can be used to quantify the effects of environmental pollutants on natural ecosystems. The ecological dose 50 percent (EcD_{50}) is the concentration of a toxicant that inhibits a microbe-mediated ecological process by 50 percent. Since microbes are an integral part of the biogeochemical cycling of elements and the flow of energy through an ecosystem, they are an important indicator of the productivity of the ecosystem. This concept is superior to the lethal dose (LD) concept because it is based on an assemblage of heterogeneous populations that are important to the ecosystem and that might be comparable to similar population assemblages of other ecosystems. The LD concept relies on the elimination of a single population that may be insignificant to the ecosystem or not comparable to other ecosystems.

Using published data, Babich et al. (1983) determined that the EcD_{50} for nitrification inhibition was 100 µg/g as soluble lead extracted from soil, based on the data of Chang and Broadbent (1982). The data of Doelman and Haanstra (1979) suggested an EcD_{50} for inhibition of respiration ranging from 0 to 7,500 µg/g total lead in soil, depending on the soil type. Peat soils showed no inhibition, sandy soils showed the most.

During decomposition, plant tissues are reduced to resistant particulate matter, as soluble organic and inorganic compounds are removed by the chemical action of soil moisture and the biochemical action of microorganisms (Odum and Drifmeyer, 1978). Each group of microorganisms specializes in the breakdown of a particular type of organic molecule. Residual waste products of one group become the food for the next group. Swift et al. (1979, p. 101) explained this relationship as a cascade effect with the following generalized pattern (Figure 8-4). Organisms capable of penetrating hard or chemically resistant plant tissue are the primary decomposers. These saprotrophs, some of which are fungi and bacteria that reside on leaf surfaces at the initial stages of senescence, produce a wide range of extracellular enzymes. Others may reside in the intestinal tract of millipedes, beetle larvae, and termites capable of mashing plant tissue into small fragments. The feces and remains of this group and the residual plant tissue are consumed by secondary decomposers, i.e., the coprophilic fungi, bacteria, and invertebrates (including protozoa) specialized for consuming bacteria. These are followed by tertiary decomposers. Microorganisms usually excrete enzymes that carry out this digestive process external to their cells. They are often protected by a thick cell coat, usually a polysaccharide. Because they are interdependent, the absence of one group in this sequence seriously affects the success of subsequent groups, as well as the rate at which plant tissue decomposes. Each group may be affected in a different way and at different lead concentrations. Lead concentrations toxic to decomposer microbes may be as low as 1 - 5 µg/g or as high as 5,000 µg/g (Doelman, 1978).

Crist et al. (1985) found no inhibition due to lead during the early stages of deciduous leaf decomposition. Green leaves were ground to a compost and innoculated with microbes from the same location. Loss of biomass was about 30 percent after 18 weeks for the controls and all lead concentrations (0 to 1,000 µg/g) of lead added as lead sulfate. The sulfate salt was considered the most common form of lead available to the decomposing leaves in the natural system. No intermediate biomass measurements were made, however. In another study, Doelman and Haanstra (1984) observed an initial inhibition of decomposition, measured by soil respiration, during the first eight weeks, followed by nearly complete recovery by about 70 weeks.

This effect was greatest for sandy and sandy loam soils, somewhat mediated in clay and sandy peat soils and virtually nonexistent in silty loam soils. No effects were observed below 1,000 µg/g. In this case, lead was added as the chloride salt.

Some studies have measured the effects of lead on specific decomposition enzymes or substrates. Haanstra and Doelman (1984) reported 50 percent inhibition (doubling of decomposition time) of glutamic acid decomposition in sandy soils at 3,500 µg/g. There was a small but distinct effect in clay soils and no effect in a calcareous silty loam soil. Frankenburger and Tabutabai (1985) measured a 5 percent inhibition of free soil amidase at about 1,000 µg/g soil. Bacterial amidase was inhibited 30 percent at about 800 mg Pb/l substrate/enzyme system.

Under conditions of mild contamination, the loss of one sensitive bacterial population may result in its replacement by a more lead-tolerant strain. Inman and Parker (1978) found that litter transplanted from a low-lead to a high-lead site decayed more slowly than high-lead litter, suggesting the presence of a lead-sensitive microorganism at the low-lead site. When high-lead litter was transplanted to the low-lead site, decomposition proceeded at a rate faster than the low-lead litter at the low-lead site. In fact, the rate was faster than the high-lead litter at the high-lead site, suggesting even the lead-tolerant strains were somewhat inhibited. The long-term effect is a change in the species composition of the ecosystem, which will be considered in greater detail in Section 8.5.3.

Delayed decomposition has been reported near smelters (Jackson and Watson, 1977), mine waste dumps (Williams et al., 1977), and roadsides (Inman and Parker, 1978). This delay is generally in the breakdown of litter from the first stage (O_1) to the second (O_2), with intact plant leaves and twigs accumulating at the soil surface. The substrate concentrations at which lead inhibits decomposition appear to be very low. Williams et al. (1977) found inhibition in 50 percent of the bacterial and fungal strains at 50 µg Pb/ml nutrient solution. The community response time for introducing lead-tolerant populations seems very fast, however. Doelman and Haanstra (1979a,b) found lead-tolerant strains had replaced non-tolerant bacteria within three years of lead exposure. These new bacteria were predominately thick-coated gram-negative strains and their effectiveness in replacing lead-sensitive strains was not evaluated in terms of soil decomposition rates.

Tyler (1982) has also shown that many species of wood-decaying fungi do not accumulate Pb, Ca, Sr, or Mn as strongly as they do other metals, even the normally toxic metal, cadmium. Accumulation was expressed as the ratio of the metal concentration in the fungus to its substrate. A ratio of greater than one implies accumulation, less than one, exclusion. Of 11 species, manganese was excluded by ten, strontium by nine, lead by eight, and calcium by seven. Potassium, at the other end of the spectrum, was not excluded by any species. The

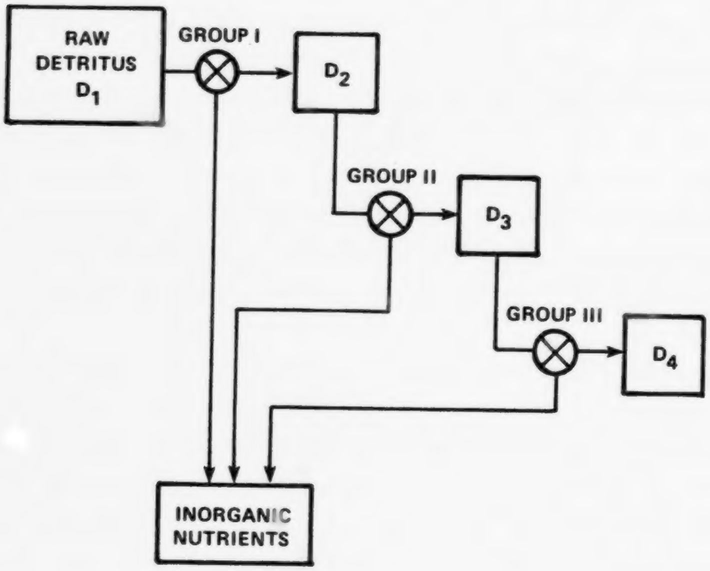

Figure 8-4. Within the decomposer food chain, detritus is progressively broken down in a sequence of steps regulated by specific groups of decomposers. Because of the cascade effect of this process, the elimination of any decomposer interrupts the supply of organic nutrients to subsequent groups and reduces the recycling of inorganic nutrients to plants. Undecomposed litter would accumulate at the stages preceding the affected decomposer.

Source: Adapted from Swift et al. (1979).

species which appeared to accumulate calcium and lead were described as having harder, less ephemeral tissues.

This relationship among calcium, strontium, and lead is consistent with the phenomenon of biopurification described in Section 8.5.2. From the data of Tyler (1982) it appears that some of the species of fungi receive lead from a source other than the nutrient medium, perhaps by direct atmospheric deposition.

8.3.2.2 Effects on Nitrifying Bacteria. The conversion of ammonia to nitrate in soil is a two-step process mediated by two genera of bacteria, Nitrosomonas and Nitrobacter. Nitrate is required by all plants, although some maintain a symbiotic relationship with nitrogen-fixing bacteria as an alternate source of nitrogen. Those that do not would be affected by a loss of free-living nitrifying bacteria, and it is known that many trace metals inhibit this nitrifying process (Liang and Tabatabai, 1977,1978). Lead is the least of these, inhibiting nitrification 14 percent at concentrations of 1,000 μg/g soil. Many metals, even the nutrient metals, manganese and iron, show greater inhibition at comparable molar concentrations. Nevertheless, soils with environmental concentrations above 1,000 µg Pb/g are frequently found. Even a 14 percent inhibition of nitrification can reduce the potential success of a plant population, as nitrate is usually the limiting nutrient in terrestrial ecosystems. In cultivated ecosystems, nitrification inhibition is not a problem if nitrate fertilizer is added to soil, but could reduce the effectiveness of ammonia fertilizer if the crops rely on nitrifying bacteria for conversion to nitrates. Rother et al. (1983) found that lead concentrations as high as 30,000 µg/g soil did not affect symbiotic nitrogen fixation in white clover (*Trifolium repens*).

8.3.2.3 Methylation by Aquatic Microorganisms. While methyllead is not a primary form of environmental lead, methylation greatly increases the toxicity of lead to aquatic organisms (Wong and Chau, 1979; Thayer and Brinckman, 1982). There is some uncertainty about whether the mechanism of methylation is biotic or abiotic. Some reports (Wong and Chau, 1979; Thompson and Crerar, 1980) conclude that lead in sediments can be methylated by bacteria. Reisinger et al. (1981) report that biomethylation of lead under aerobic or anaerobic conditions does not occur and such reports are probably due to sulfide-induced chemical conversion of organic lead salts. These authors generally agree that tetramethyl lead can be formed under environmental conditions when another tetravalent organolead compound is available, but methylation of divalent lead salts such as $Pb(NO_3)_2$ does not appear to be significant. Jarvie et al. (1983) also report that they were unable to produce any definite evidence for biomethylation of lead.

8.3.2.4 Summary of Effects on Microorganisms. It appears that microorganisms are more sensitive than plants to soil lead pollution and that changes in the composition of bacterial populations may be an early indication of lead effects. Delayed decomposition may occur at 750 µg Pb/g soil and nitrification inhibition at 1,000 µg/g. Many of the environmental variables that can raise or lower these estimates are not yet known. In certain chemical environments, the highly toxic tetramethyllead can be formed, but this process does not appear to be mediated by aquatic microorganisms.

8.4 EFFECTS OF LEAD ON DOMESTIC AND WILD ANIMALS

8.4.1 Vertebrates

8.4.1.1 Terrestrial Vertebrates. Forbes and Sanderson (1978) have reviewed reports of lead toxicity in domestic and wild animals. Lethal toxicity can usually be traced to consumption of lead battery casings, lead-based paints, oil wastes, putty, linoleum, pesticides, lead shot, or forage near smelters. Except for lead shot ingestion, these problems can be solved by proper management of domestic animals. However, the 3,000 tons of lead shot falling annually along waterways and other hunting grounds continues to be a problem.

A single pellet of lead shot weighs about 110 mg, and 70 percent of this may be eroded in ringed turtle dove gizzards over a period of 14 days (Kendall et al., 1982). Their data showed an immediate elevation of blood lead and reduction of aminolevulinic acid dehydrogenase (ALA-D) activity within one day of swallowing two pellets. Feierabend (1983) reviewed 97 reports on the effects of lead shot on waterfowl. Of the estimated 80 to 125 million waterfowl in North America, 1.5 to 38 million die each year from lead poisoning. Many more are greatly impaired by chronic sublethal exposure. Reichell et al. (1984) reported that 17 of 293 bald eagles sampled had lead concentrations in their liver high enough to suspect lead poisoning. The 293 specimens were found dead or nearly dead during 1978 to 1983. The most common causes of death were trauma from being hit by a motor vehicle (20 percent) and shooting (19 percent). Bjorn et al., (1982) also reported the uptake of lead shot by grazing cattle near a trapshooting site.

Bull et al. (1983) and Osborn et al. (1983) reported extensive bird mortality that could be attributed to alkyl lead pollution of the Mersey Estuary in the United Kingdom. Bull et al. (1983) found 3-18 µg/g alkyl lead in dead birds, 1-14 µg/g in sick birds, and 0.3-1.2 µg/g in apparently healthy birds. Osborn et al. (1983), in laboratory studies, found that 2000 µg/day alkyl lead in the diet caused heavy mortality and 200 µg/day caused tremors, impaired balance, and feeding irregularities, although no mortality was observed. Tissue concentrations of alkyl lead at the lower dose were in the range of 0.2 to 5.4 µg/g. The authors concluded that many of the apparently healthy wild birds were experiencing symptoms likely to impair their chances for survival.

Awareness of the routes of uptake is important in interpreting the exposure and accumulation in vertebrates. Inhalation rarely accounts for more than 10 - 15 percent of the daily intake of lead (National Academy of Sciences, 1980). Much of the inhaled lead is trapped on the walls of the bronchial tubes and passes to the stomach embedded in swallowed mucus. Because lead concentrations in lakes or running stream water are quite low, intake from drinking water may also be insignificant unless the animal drinks from a stagnant or otherwise contaminated source.

Food is the largest contributor of lead to animals. The type of food an herbivore eats determines the rate of lead ingestion. More than 90 percent of the total lead in leaves and bark may be due to surface deposition, but relatively little surface deposition may be found on some fruits, berries, and seeds that have short exposure times. Roots intrinsically have no surface deposition. Similarly, ingestion of lead by a carnivore depends mostly on deposition on herbivore fur and somewhat less on lead in herbivore tissue. Harrison and Dyer (1984) estimated that mule deer grazing in the Rocky Mountain National Park would exceed acceptable lead exposure by grazing on roadside vegetation for just 1 to 2 percent of the time. This estimate was based on the assumption that the upper limit of exposure should be 3,000 µg Pb/day. Mule deer grazing on non-roadside forage would consume about 1,500 µg/day.

The type of food eaten is a major determinant of lead body burdens in small mammals. Goldsmith and Scanlon (1977) and Scanlon (1979) measured higher lead concentrations in insectivorous species than in herbivorous species, confirming the earlier work of Quarles et al. (1974), which showed body burdens of granivores < herbivores < insectivores, and Jeffries and French (1972) that granivores < herbivores. Animals in these studies were analyzed whole minus the digestive tract. It is likely that observed diet-related differences were somewhat diluted by including fur in the analysis, because fur lead might be similar for small mammals from the same habitats with different feeding habits.

Since 1977, there has been a trend away from whole body analyses toward analyses of isolated tissues, especially bones and blood. Bone concentrations of lead are better than blood as indicators of long-term exposure. Because natural levels of blood lead are not well known for animals and blood is not a good indicator of chronic exposure, blood lead is poorly suited for estimating total body burdens. One experiment with sheep shows the rapid response of blood to changes in lead ingestion and the relative contribution of food and air to the total blood level. Ward et al. (1978) analyzed the blood in sheep grazing near a highway (0.9 µg/g ml) and in an uncontaminated area (0.2 µg/ml). When sheep from the uncontaminated area were allowed to graze near the roadway, their blood levels rose rapidly (within 1 day) to about 3.0 µg/ml, then decreased to 2.0 µg/ml during the next 2 days, remaining constant for the remainder of the 14-day period. Sheep from the contaminated area were moved to the uncontaminated area, where upon their blood dropped to 0.5 µg/ml in 10 days and decreased to 0.3 µg/ml during the next 180 days. Sheep in the uncontaminated area that were fed forage from the roadside experienced an increase in blood lead from 0.2 to 1.1 µg/ml in 9 days. Conversely, sheep from the uncontaminated area moved to the roadside but fed forage only from the uncontaminated site experienced an increase from 0.2 to 0.5 µg/ml in 4 days. These data show that both air and food contribute to lead in blood and that blood lead concentrations are a function of both the recent history of lead exposure and the long-term storage of lead in bone tissue.

Beyer et al. (1985) reported a decrease in red blood cell ALA-D activity for 14 small mammals and 15 songbirds in a habitat near a smelter. There were no changes in packed cell volume or hemoglobin concentrations and little evidence of gross or microscopic lesions that could be attributed to metal poisoning. Intranuclear inclusion bodies were found in a kidney of one shrew. The soil concentrations at the surface were 1,200-2,700 µg/g. Foliage concentrations were 21 µg/g and the fruits and berries averaged 4 µg/g, a typical pattern for the distribution of lead of atmospheric origin. The authors attributed the relatively minor effects of lead on the animals to the fact that the mice, shrews, and songbirds were eating primarily fruits and berries, not leaves. They also considered the possibility that populations of some species had been previously reduced or eliminated by the emissions from the smelter.

Chmiel and Harrison (1981) showed that, for small mammals, the highest concentrations of lead occurred in the bones (Table 8-2), with kidney and liver concentrations somewhat less. They also showed greater bone concentrations in insectivores than herbivores, both at the control and contaminated sites. Clark (1979) found lead concentrations in shrews, voles, and brown bats from roadside habitats near Washington, D.C., to be higher than any previously reported. His estimates of dosages (7.4 mg Pb/kg·day) exceed those that normally cause mortality or reproductive impairment in domestic mammals (1.5-9 mg Pb/g·day) (Hammond and Aronson, 1964; James et al., 1966; Kelliher et al., 1973). Traffic density was the same as reported by Chmiel and Harrison (1981), nearly twice that of Goldsmith and Scanlon (1977) (Table 8-2). The body lead burden of shrews exceeded mice, which exceeded voles. Beresford et al. (1981) found higher lead in box turtles within 500 m of a lead smelter than in those from control sites. Bone lead exceeded kidney and liver lead as in small mammals.

Kusseberth et al. (1984) reported that lead in the bones of small mammals indigenous to a habitat near a battery reclamation plant decreased exponentially with distance from the battery plant. The observed pattern was similar to that reported for lead in roadside soils and vegetation reported in Section 7.2.2.1.1. They also reported findings of intranuclear inclusions in renal tubular epithelial tissue in one vole and four deer mice.

There are few studies reporting lead in vertebrate tissues from remote sites. Elias et al. (1976, 1982) reported tissue concentrations in voles, shrews, chipmunks, tree squirrels, and pine martens from the remote High Sierra. Bone concentrations were generally only 2 percent of those reported from roadside studies and 10 percent of the controls of roadside studies (Table 8-2), indicating the roadside controls were themselves contaminated to a large degree. Furthermore, biogeochemical calculations suggest that even animals in remote areas have bone lead concentrations 50 to 500 times natural background levels. The natural concentration of lead in the bones of herbivores is about 0.04 ng/g dry weight (Table 8-1). This value may vary regionally with geochemical anomalies in crustal rock, but provides a reasonable indicator of contamination. Natural levels of lead in carnivore bone tissue should be somewhat lower, with omnivores generally in between (Elias and Patterson, 1980; Elias et al., 1982).

Table 8-2 shows the results of several studies of small animal bone tissue. To convert reported values to a common basis, assumptions were made of the average water content, calcium

TABLE 8-2. ESTIMATES OF THE DEGREE OF CONTAMINATION OF HERBIVORES, OMNIVORES, AND CARNIVORES

Data are based on published concentrations of lead in bone tissue (corrected to dry weight as indicated). Degree of contamination is calculated as observed/natural Pb. Natural lead concentrations are from Table 8-1. Concentrations are in µg Pb/g dw.

Organism	Bone Pb conc.	Ref.	Estimated degree of contamination bone
Herbivores			
Vole-roadside	38	1	320
Vole-roadside	17	2	140
-control	5	2	42
Vole-orchard	73	5	610
-control	9	5	75
Vole-remote	2	11	17
Deer mouse-roadside	25	2	210
-control	5.7	2	48
Deer mouse-near battery plant	80	13	650
-control	2	13	18
Deer mouse-roadside	29	3	240
-control	7.2	3	60
Deer mouse-roadside	52	4	430
-control	5	4	42
Mouse-roadside	19	2	160
-control	9.3	2	78
Mouse-roadside	109	2	910
-control	18	2	150
Average herbivore			
roadside (7)	41		340
control (7)	8.5		71
remote (2)	2		17
Omnivores/frugivores			
Woodmouse-roadside	67	1	840
-control	25	1	310
Composite-roadside	22	7	280
-control	3	7	37
Chipmunk-remote	2	1	25
Tree squirrel-remote	1.3	11	16
Feral pigeon-urban	670	6	8400
-rural	5.7	6	71
Feral pigeon-urban	250	12	3100
-suburan	33	12	410
-rural	12	12	150

(continued)

TABLE 8-2. (continued)

Organism	Bone Pb conc.	Ref.	Estimated degree of contamination bone
Starling-roadside	210	7	2600
-control	13	7	160
Robin-roadside	130	7	1600
-control	41	7	510
Sparrow-roadside	130	7	1600
-control	17	7	200
Blackbird-roadside	90	7	1100
-control	7	7	88
Grackle-roadside	63	7	790
-control	22	7	280
Rats-roadside	310[a]	9	10000
-control	15[a]	9	500
Average omnivore			
roadside (7)	102		1260
urban (1)	670		8400
control (7)	18		230
remote (2)	1.7		21
Carnivores			
Box turtle-smelter	91[a]	8	3000
-control	5.7[a]	8	190
Egret-rural	12[a]	10	400
Gull-rural	11[a]	10	370
Mink-rural	1.5	14	50
Shrew-roadside	67	2	2200
-control	12	2	400
Shrew-roadside	193	1	6400
-control	41	1	1400
Shrew-remote	4.6	1	150
Pine marten-remote	1.4	11	47
Average carnivore			
roadside (3)	190		6200
smelter (1)	91		3000
rural (2)	11		385
control (4)	18		620
remote (2)	3		99

[a] Dry weight calculated from published fresh weights assuming 35 percent water.

1. Chmiel and Harrison, 1981
2. Getz et al., 1977b
3. Welch and Dick, 1975
4. Mierau and Favara, 1975
5. Elfving et al., 1978
6. Hutton and Goodman, 1980
7. Getz et al., 1977a
8. Beresford et al., 1981
9. Mouw et al., 1975
10. Hulse et al., 1980
11. Elias et al., 1982
12. Johnson et al., 1982b
13. Kisseberth et al., 1984
14. Ogle et al., 1985

concentration, and average crustal concentration. Because ranges of natural concentrations of lead in bones, plants, soils, and air are known with reasonable certainty (Table 8-1), it is possible to estimate the degree of contamination of vertebrates from a wide range of habitats. It is important to recognize that these are merely estimates that do not allow for possible errors in analysis or anomalies in regional crustal abundances of lead.

8.4.1.2 Effects on Aquatic Vertebrates. Lowe et al. (1985) reported the results of a nationwide survey of metal concentrations in freshwater fish during the period 1979 to 1981. At 112 monitoring stations they found an average (geometric mean) of 0.19 µg Pb/g wet weight for the period 1978-79 and 0.17 µg/g for 1980-81. Several laboratories have reported experiments that measure the effects of lead on freshwater fish. Two requirements limit the evaluation of literature reports of lead effects on aquatic organisms. First, any laboratory study should incorporate the entire life cycle of the organism studied. It is clear that certain stages of a life cycle are more vulnerable than others (Hodson, 1979, Hodson et al., 1979). For fish, the egg or fry is usually most sensitive. Secondly, the same index must be used to compare results. Christensen et al. (1977) proposed three indices useful for identifying the effects of lead on organisms. A molecular index reports the maximum concentration of lead causing no significant biochemical change; residue index is the maximum concentration showing no continuing increase of deposition in tissue; and a bioassay index is the maximum concentration causing no mortality, growth change, or physical deformity. These indices are comparable to those of physiological dysfunction (molecular, tissue, and organismic) discussed in Section 8.1.3.

From the standpoint of environmental protection, the most useful index is the molecular index. This index is comparable to the point of initial response discussed previously and is equivalent to the "safe concentration" originally described by the U.S. Environmental Protection Agency (Battelle, 1971) as being the concentration that permits normal reproduction, growth, and all other life-processes of all organisms. It is unfortunate that very few of the toxicity studies in the aquatic literature report safe concentrations as defined above. Nearly all report levels at which some or all of the organisms die.

Hematological and neurological responses are the most commonly reported effects of extended lead exposures in aquatic vertebrates. Hematological effects include the disabling and destruction of mature red blood cells and the inhibition of the enzyme ALA-D required for hemoglobin synthesis. At low exposures, fish compensate by forming additional red blood cells. These red blood cells often do not reach maturity. At higher exposures, the fish become anemic. Symptoms of neurological responses are difficult to detect at low exposure, but higher exposure can induce neuromuscular distortion, anorexia, and muscle tremors. Spinal curvature eventually occurs with time or increased concentration (Hodson 1979; Hodson et al., 1977). Weis and Weis (1982) found spinal curvature in developing eggs of killifish when the embryos had been exposed to 10 μg Pb/ml during the first 7 days after fertilization. All batches showed some measure of curvature, but those that were most resistant to lead were least resistant to the effects of methylmercury. Sippel et al. (1983) reported that black fin and spinal curvature in rainbow trout were the most reliable clinical tests for lead toxicity at low levels. These effects appear at about 120 μg/l before effects on red blood cells, liver function, or histopathological indications in the liver, spleen, kidneys, gills, brain, spinal cord, or gastrointestinal tract.

The biochemical changes used by Christensen et al. (1977) to determine the molecular index for brook trout were 1) increases in plasma sodium and chloride and 2) decreases in glutamic oxalacetic transaminase activity and hemoglobin. They observed effects at 0.5 μg/l, which is 20-fold less than the lower range (10 μg/l) suggested by Wong et al. (1978) to cause significant detrimental effects. Hodson et al. (1978a) found tissue accumulation and blood parameter changes in rainbow trout at 13 μg/l. This was the lowest experimental level, and only slightly above the controls, which averaged 4 μg/l. They concluded, however, that because spinal curvature does not occur until exposures reach 120 μg/l, rainbow trout are adequately protected at 25 μg/l.

Aside from the biochemical responses discussed by Christensen et al. (1977), the lowest reported exposure concentration that causes hematological or neurological effects is 8 μg/l (Hodson, 1979). Christensen's group dealt with subcellular responses, whereas Hodson's group dealt primarily with responses at the cellular or higher level. Hodson et al. (1978a) also reported that lead in food is not available for assimilation by fish, that most of their lead comes from water, and that decreasing the pH of water (as in acid rain) increases the uptake of lead by fish (Hodson et al., 1978b). Patrick and Loutit (1978), however, reported that tissue lead in fish reflects the lead in food if the fish are exposed to the food for more than a few days. Hodson et al. (1980) also reported that, although the symptoms are similar (spinal deformation), lead toxicity and ascorbic acid deficiency are not metabolically related.

8.4.2 Invertebrates

Insects have lead concentrations that correspond to those found in their habitat and diet. Herbivorous invertebrates have lower concentrations than do predatory types (Wade et al., 1980). Among the herbivorous groups, sucking insects have lower lead concentrations than chewing insects, especially in regions near roadsides, where more lead is found on the surfaces of vegetation. Williamson and Evans (1972) found gradients away from roadsides are not the same as with vertebrates, in that invertebrate lead decreases more slowly than vertebrate lead relative to decreases in soil lead. They also found great differences between major groups of invertebrates. Wood lice in the same habitat, eating the same food, had eight times more lead than millipedes.

There are a few isolated reports on the effects of lead on the physiology of insects. Hopkin and Martin (1984) fed hepatopancreas tissue from the woodlouse *Oniscus asellus* L. to centipedes (*Lithobius variegatus*) and found that the lead was not assimilated by the centipede but passed directly through the midgut within four days of consumption. The centipedes were fed 1 to 16 µg Pb. Bengtsson et al. (1983) observed delayed growth in populations of *Onychiurus armatus* (Tullb.), a soil insect that feeds on detritus and microorganisms. The insects were fed a diet of fungi that had been grown on media from 0 to 150 µg Pb/g and had accumulated mycelial concentrations of 8 to 3100 µg/g in direct proportion to media concentrations. The F1 and F2 generations experienced a marked decline in growth rate, measured as length versus age, but eventually acheived the same maximum length as the controls. Lead was stored for the first two weeks during the life cycle, then excreted. The reduction in growth rate, or delay in achieving maximum length, was seen to be significant to the reproductive process because the length at first egg-laying appeared to be relatively constant at 0.90 to 0.97 mm. This evidence suggests that reduced growth rate might be accompanied by delayed sexual maturity.

The distribution of lead among terrestrial gastropod tissues was reported by Ireland (1979). He found little difference among the foot, skin, mantle, digestive gland, gonad, and intestine. There are no reports of lead toxicity in soil invertebrates. In a feeding experiment, however, Coughtrey et al. (1980) found decreased tolerance for lead by microorganisms from the guts of insects at 800 µg Pb/g food. Many roadside soils fall in this range.

In *Cepaea hortensis*, a terrestrial snail, Williamson (1979) found most of the lead in the digestive gland and gonadal tissue. He also determined that these snails can lose 93 percent of their whole body lead burden in 20 days when fed a low-lead diet in the laboratory. Since no analyses of the shell were reported, elimination of lead from this tissue cannot be evaluated. A continuation of the study (Williamson, 1980) showed that body weight, age, and day-length influenced the lead concentrations in soft tissues.

Beeby and Eaves (1983) addressed the question of whether uptake of lead in the garden snail, *Helix aspersa*, is related to the nutrient requirement for calcium during shell formation and reproductive activity. They found concentrations of both metals were strongly correlated with changes in dry weight and little evidence for correlation of lead with calcium independent of weight gain or loss. Lead in the diet remained constant.

Gish and Christensen (1973) found lead in whole earthworms to be correlated with soil lead, with little rejection of lead by earthworms. Consequently, animals feeding on earthworms from high-lead soils might receive toxic amounts of lead in their diets, although there was no evidence of toxic effects on the earthworms (Ireland, 1977). Ash and Lee (1980) cleared the digestive tracts of earthworms and still found direct correlation of lead in earthworms with soil lead; in this case, soil lead was inferred from fecal analyses. These authors found differences among species of earthworms. Ireland and Richards (1977) also found species differences in earthworms, as well as some localization of lead in subcellular organelles of chloragogue and intestinal tissue. In view of the fact that chloragocytes are believed to be involved with waste storage and glycogen synthesis, the authors concluded that this tissue is used to sequester lead in the manner of vertebrate livers. Species differences in whole body lead concentrations could not be attributed to selective feeding or differential absorption, unless the differential absorption occurs only at elevated lead concentrations. The authors suggested that the two species have different maximum tolerances for body lead but gave no indication of physiological dysfunction when the maximum tolerance was reached. In soils with a total lead concentration of 1,800 µg/g dry weight (Ireland, 1975), Lumbricus rubellus had a whole body concentration of 3,600 µg/g, while Dendrobaene rubida accumulated 7,600 µg/g in the same location (Ireland and Richards, 1977). Because this difference was not observed at the control site (15 µg/g soil), it can be assumed that at some soil concentration between 15 and 1,800 µg/g, different species of earthworms begin to accumulate different amounts of lead. The authors concluded that D. rubida can simply tolerate higher tissue lead concentrations, implying that soil concentrations of 1,800 µg/g are toxic to L. rubellus. This concentration would be considerably lower than soil lead concentrations that cause effects in plants, and similar to that which can affect soil microorganisms. Ma et al. (1983) found that the uptake of lead by populations of earthworms near a zinc smelter complex was related to soil pH and organic content. In the observed range of 3.5 to 6.1, low soil pH increased the accumulation of lead by L. rubellus. Likewise, for the range of 2.2 to 8.6 percent organic matter, earthworms accumulated more lead when exposed to soil at the lower end of the range. Kruse and Barrett (1985) measured greater lead concentrations in cleared earthworms from sludge-treated soils. The sludge amended soil was 1.5 times the lead content of the control soil, and the corresponding earthworms were about 3.5 times higher.

Aquatic insects appear to be resistant to high levels of lead in water. To be conclusive, toxicity studies must observe invertebrates through an entire life cycle, although this is infrequently done. Anderson et al. (1980) found LC_{50}'s for eggs and larvae of Tanytarsus dissimilis, a chironomid, to be 260 µg/l. This value is 13 - 250 times lower than previously reported by Warnick and Bell (1969), Rehwoldt et al. (1973), and Nehring (1976). However, Spehar et al. (1978) found that mature amphipods (Gammarus pseudolimnaeus) responded negatively to lead at 32 µg/l. Fraser et al. (1978) found that adult populations of a freshwater isopod (Asellus aquaticus) have apparently developed a genetic tolerance for lead in river sediments.

Newman and McIntosh (1982) investigated freshwater gastropods, both grazing and burrowing. Lead concentrations in the grazers (Physa integra, Pseudosuccinea columella, and Helisoma trivolvis) were more closely correlated with water concentrations than with lead in the food. Lead in the burrowing species, Campeloma decisum, was not correlated with any environmental factor. These authors (Newman and McIntosh, 1983) also reported that both Physa integra and Campeloma decisum are able to eliminate lead from their soft tissue when transferred to a low-lead medium, but that tissue lead stabilized at a level higher than found in populations living permanently in the low-lead environment. This would seem to indicate the presence of a persistent reservoir of lead in the soft tissues of these gastropods. Tessier et al. (1984) measured metal accumulation in the tissues of the freshwater bivalve Elliptio complanata and concluded that concentrations of lead in the bivalve were directly related to concentrations of lead in that fraction of the sediment that can be most easily extracted. The highest concentrations of lead were in the gills, mantle, and hepatopancreatic tissue. They concluded further that lead may enter the organism through the gills more so than through the digestive tract, and that the presence of amorphous iron oxyhydroxides reduces the concentration of metals in the bivalve tissues by selectively competing for the binding sites.

Everard and Denny (1984) observed that freshwater snails (Lymnaea peregra) accumulate lead in their digestive glands, feet, and shells when fed a diet enriched with lead. These snails are efficient grazers of Aufwuchs, the epiflora and epifauna that coat all submerged surfaces of the euphotic zone. Granular bodies, thought to be precipitated lead phosphate, were observed in the gut epithelium, gut lumen, digestive gland, and the foot of those snails fed a lead-rich diet, but not in the controls. Snails transferred from a lead-contaminated environment to a lead-free environment could be cleared of lead in their soft tissues in about four weeks, but the concentration of lead in the shells did not decrease. Borgmann et al. (1978) found increased mortality in a freshwater snail, Lymnaea palutris, associated with stream water with a lead content as low as 19 μg/l. Full life cycles were studied to estimate population productivity. Although individual growth rates were not affected, increased mortality, especially at the egg hatching stage, effectively reduced total biomass production at the population level. Production was 50 percent at 36 μg/l and 0 percent at 48 μg Pb/l.

The relationship between LC_{50} and initial physiological response is not immediately obvious. It is certain that some individuals of a population experience physiological dysfunction at concentrations well below that where half of them die. For example, Biesinger and Christensen (1972) observed minimum reproductive impairment in Daphnia at 6 percent of the LC_{50} (450 μg/l) for this species.

8.4.3 Summary of Effects on Animals

While it is impossible to establish a safe limit of daily lead consumption, it is reasonable to generalize that a regular diet of 2-8 mg Pb/kg·day body weight over an extended period of time (Botts, 1977) will cause death in most animals. Animals of the grazing food chain are affected most directly by the accumulation of aerosol particles on vegetation surfaces and somewhat indirectly by the uptake of lead through plant roots. Many of these animals consume more than 1 mg Pb/kg·day in habitats near smelters and roadsides, but no toxic effects have been documented. Animals of the decomposer food chain are affected indirectly by lead in soil which can eliminate populations of microorganisms preceding animals in the food chain or occupying the digestive tract of animals and aiding in the breakdown of organic matter. Invertebrates may also accumultate lead at levels toxic to their predators.

Aquatic animals are affected by lead at water concentrations lower than previously considered safe (50 μg Pb/l) for wildlife. These concentrations occur commonly, but the contribution of atmospheric lead to specific sites of high aquatic lead is not clear.

8.5 EFFECTS OF LEAD ON ECOSYSTEMS

There is wide variation in the mass transfer of lead from the atmosphere to terrestrial ecosystems. Even within the somewhat artificial classification of undisturbed, cultivated, and urban ecosystems, reported fluxes in undisturbed ecosystems vary by nearly 20-fold. Smith and Siccama (1981) report 270 g/ha·yr in the Hubbard Brook forest of New Hampshire; Lindberg and Harriss (1981) found 150 g/ha·yr in the Walker Branch watershed of Tennessee; and Elias et al. (1976) found 15 g/ha·yr in a remote subalpine ecosystem of California. Jackson and Watson (1977) found 1,000,000 g/ha·yr near a smelter in southeastern Missouri. Getz et al. (1977c) estimated 240 g/ha·yr by wet precipitation alone in a rural ecosystem largely cultivated and 770 g/ha·yr in an urban ecosystem.

One factor causing great variation is remoteness from source, which translates to lower air concentrations, smaller particles, and greater dependence on wind as a mechanism of deposition (Elias and Davidson, 1980). Another factor is type of vegetation cover. Deciduous leaves may, by the nature of their surface and orientation in the wind stream, be more suitable deposition surfaces than conifer needles. Davidson et al. (1982) discussed the influence of leaf surface on deposition rates to grasses.

The history of lead contamination in roadside ecosystems has been reviewed by Smith (1976). Recent studies have shown three areas of concern where the effects of lead on ecosystems may be extremely sensitive (Martin and Coughtrey, 1981; Smith, 1981). First, decomposition is delayed by lead, as some decomposer microorganisms and invertebrates are inhibited by soil lead. Secondly, the natural processes of calcium biopurification are circumvented by the accumulation of lead on the surfaces of vegetation and in the soil reservoir. Thirdly, some ecosystems experience subtle shifts toward lead tolerant plant populations. These problems all arise because lead in ecosystems is deposited on vegetation surfaces, accumulates in the soil reservoir, and is not removed with the surface and ground water passing out of the ecosystem. Other potential effects are discussed that may occur because of the long-term build-up of lead in soil.

8.5.1 Delayed Decomposition

The flow of energy through an ecosystem is regulated largely by the ability of organisms to trap energy in the form of sunlight and to convert this energy from one chemical form to another (photosynthesis). Through photosynthesis, plants convert light to stored chemical energy. Starch is only a minor product of this energy conversion. The most abundant substance produced by net primary production is cellulose, a structural carbohydrate of plants. Terrestrial ecosystems, especially forests, accumulate a tremendous amount of cellulose as woody tissue of trees. Few animals can digest cellulose and most of these require symbiotic associations with specialized bacteria. It is no surprise then, that most of this cellulose must eventually pass through the decomposer food chain. Litter fall is the major route for this pathway. Because 80 percent or more of net primary production passes through the decomposing food chain (Swift et al., 1979), the energy of this litter is vital to the rest of the plant community and the inorganic nutrients are vital to plants.

The amount of lead that causes litter to be resistant to decomposition is not known. Although laboratory studies show that 50 µg Pb/ml nutrient medium definitely inhibits soil bacterial populations, field studies indicate little or no effect at 600 µg/g litter (Doelman and Haanstra, 1979b). One explanation is that the lead in the laboratory nutrient medium was readily available, while the lead in the litter was chemically bound to soil organic matter. Indeed, Doelman and Haanstra (1979a) demonstrated the effects of soil lead content on delayed decomposition: sandy soils lacking organic complexing compounds showed a 30 percent inhibition of decomposition at 750 µg/g, including the complete loss of major bacterial species, whereas the effect was reduced in clay soils and non-existent in peat soils. Organic matter maintains the cation exchange capacity of soils. A reduction in decomposition rate was observed by Doelman and Haanstra (1979a) even at the lowest experimental concentration of lead, leading to the conclusion that some effect might have occurred at even lower concentrations.

When decomposition is delayed, nutrients may be limiting to plants. In tropical regions or areas with sandy soils, rapid turnover of nutrients is essential for the success of the forest community. Even in a mixed deciduous forest, a significant portion of the nutrients, especially nitrogen and sulfur, may be found in the litter reservoir (Likens et al. 1977). Annual litter inputs of calcium and nitrogen to the soil account for about 60 percent of root uptake. With delayed decomposition, plants must rely on precipitation and soil weathering for the bulk of their nutrients. Furthermore, the organic content of soil may decrease, reducing the cation exchange capacity of soil.

8.5.2 Circumvention of Calcium Biopurification

Biopurification is a process that regulates the relative concentrations of nutrient to non-nutrient elements in biological components of a food chain. In the absence of absolute knowledge of natural lead concentrations, biopurification can be a convenient method for estimating the degree of contamination. Following the suggestion by Comar (1965) that carnivorous animals show reduced Sr/Ca ratios compared to herbivorous animals which, in turn show less than plants, Elias et al. (1976, 1982) developed a theory of biopurification, which hypothesizes that calcium reservoirs are progressively purified of Sr, Ba, and Pb in successive stages of a food chain. In other words, if the Sr/Ca and Ba/Ca ratios are known, the natural Pb/Ca ratio can be predicted and the observed Pb/Ca to natural Pb/Ca ratio is an expression of the degree of contamination. Elias et al. (1976, 1982) and Elias and Patterson (1980) observed continuous biopurification of calcium in grazing and detrital food chains by the progressive exclusion of Sr, Ba, and Pb (Figure 8-5). It is now believed that members of grazing and decomposer food chains are contaminated by factors of 30 - 500, i.e., that 97 - 99.9 percent of the lead in organisms is of anthropogenic origin. Burnett and Patterson (1980) have shown a similar pattern for a marine food chain.

The mechanism of biopurification relies heavily on the selective transport of calcium across membranes, the selective retention of non-nutrients at physiologically inactive binding sites, and the reduced solubility of non-nutrient elements in the nutrient medium of plants and animals. For example, lead is bound more vigorously to soil organic complexes and is less soluble in soil moisture (Section 6.5.1). Lead is also adsorbed to cell walls in the root apoplast, is excluded by the cortical cell membrane, and is isolated as a precipitate in subcellular vesicles of cortical cells (Koeppe, 1981). Further selectivity at the endodermis results in a nutrient solution of calcium in the vascular tissue that is greatly purified of lead. Similar mechanisms occur in the stems and leaves of plants, in the digestive and circulatory systems of herbivores and carnivores, and in the nutrient processing mechanisms of insects.

Atmospheric lead circumvents the natural biopurification of calcium. Deposition on plant surfaces, which accounts for 90 percent of the total plant lead, increases the ratio of Pb/Ca in the diet of herbivores. Deposition on herbivore fur increases the Pb/Ca ratio in the diet

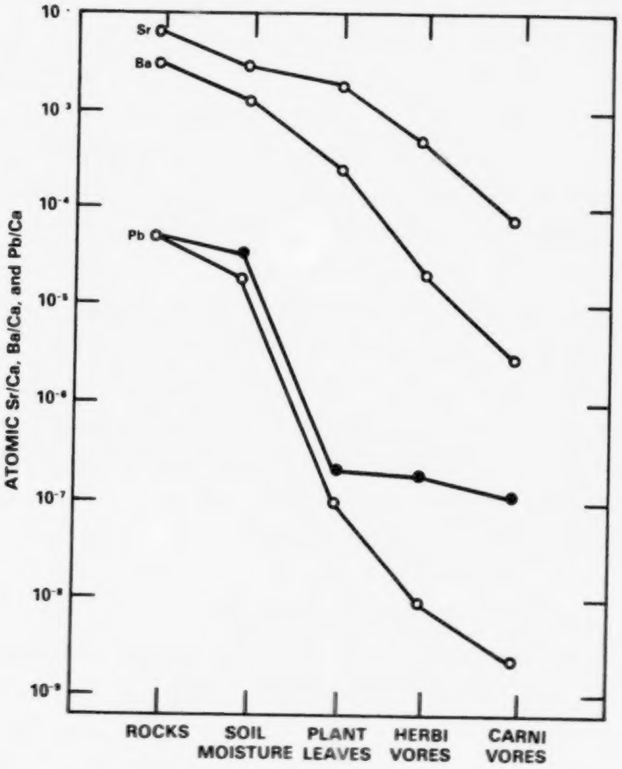

Figure 8-5. The atomic ratios Sr/Ca, Ba/Ca and Pb/Ca (○) normally decrease by several orders of magnitude from the crustal rock to ultimate carnivores in grazer and decomposer food chains. Anthropogenic lead in soil moisture and on the surfaces of vegetation and animal fur interrupt this process to cause elevated Pb/Ca ratios (●) at each stage of the sequence. The degree of contamination is the ratio of Total Pb/Ca *vs.* Natural Pb/Ca at any stage. Ba/Ca and Sr/Ca ratios are approximate guidelines to the expected natural Pb/Ca ratio.

Source: Adapted from Elias et al. (1982).

of carnivores. Atmospheric lead consumed by inhalation or grooming, possibly 15 percent of the total intake of lead, represents sources of lead that were non-existent in prehistoric times and therefore were not present in the food chain.

8.5.3 Population Shifts Toward Lead-Tolerant Populations

It has been observed that plant communities near smelter sites are composed mostly of lead tolerant plant populations (Antonovics et al., 1971). In some cases, these populations appear to have adapted to high-lead soils, since populations of the same species from low-lead soils often do not thrive on high-lead soils (Jowett, 1964). Similar effects have been observed for soils enriched to 28,000 μg/g dry weight with ore lead (Høiland and Oftedal, 1980) and near roadsides at soil concentrations of 1,300 μg/g dry weight (Atkins et al., 1982). In these situations, it is clear that soil lead concentration has become the dominant factor in determining the success of plant populations and the stability of the ecological community. Soil moisture, soil pH, light intensity, photoperiod, and temperature are all secondary factors (Antonovics et al., 1971). Strategies for efficient use of light and water, and for protection from temperature extremes, are obliterated by the succession of lead-tolerant plant populations. Smith and Bradshaw (1972) concluded that lead-tolerant plant populations of *Festuca rubra* and *Agrostis tenuis* can be used to stabilize toxic mine wastes with lead concentrations as high as 80,000 μg/g.

8.5.4 Biogeochemical Distribution of Lead in Ecosystems

Inputs of natural lead to ecosystems, approximately 90 percent from rock weathering and 10 percent from atmospheric sources, account for slightly more than the hydrologic lead outputs in most watersheds (Patterson, 1980). The difference is small and accumulation in the ecosystem is significant only over a period of several thousand years. In modern ecosystems, with atmospheric inputs exceeding weathering by factors of 10 - 1000, greater accumulation occurs in soils. This reservoir must be treated as lacking a steady state condition (Heinrichs and Mayer, 1977, 1980; Siccama and Smith, 1978). Odum and Drifmeyer (1978) describe the role of detrital particles in retaining a wide variety of pollutant substances, and this role may be extended to include non-nutrient substances.

It appears that plant communities have a built-in mechanism for purifying their own nutrient medium. As a plant community matures through successional stages, the soil profile develops a stratified arrangement that retains a layer of organic material near the surface. This organic layer becomes a natural site for the accumulation of lead and other non-nutrient metals that might otherwise interfere with the uptake and utilization of nutrient metals. But the rate accumulation of lead in this reservoir may eventually exceed the capacity of the reservoir. Johnson et al. (1982a) have established a baseline of 80 stations in forests of the northeast United States. In the litter component of the forest floor, they measured an average lead concentration of 150 µg/g. Near a smelter, they measured 700 µg/g and near a highway, 440 µg/g. They presented some evidence from buried litter that predevelopment concentrations were 24 µg/g. On an area basis, the present concentrations range from 0.7 to 1.8 g Pb/m^2. Inputs of 270 g/ha·yr measured in the Hubbard Brook forest would account for 1.0 g Pb/m^2 in forty years if all of the lead were retained. The 80 stations will be monitored regularly to show temporal changes. Evidence for recent changes in litter lead concentrations is documented in the linear relationship between forest floor lead concentration and age of forest floor, up to 100 years.

Lead in the detrital reservoir is determined by the continued input of atmospheric lead from the litter layer, the passage of detritus through the decomposer food chain, and the rate of leaching into soil moisture. There is strong evidence that soil has a finite capacity to retain lead (Zimdahl and Skogerboe, 1977). Harrison et al. (1981) observed that most of the lead in roadside soils above 200 µg/g is found on Fe-Mn oxide films or as soluble lead carbonate. Elias et al. (1982) have shown that soil moisture lead is derived from the leachable/organic fraction of soil, not the inorganic fraction. Lead is removed from the detrital reservoir by the digestion of organic particles in the detrital food chain and by the release of lead to soil moisture. Both mechanisms result in a redistribution of lead among all of the reservoirs of the ecosystem at a very slow rate. A closer look at the mechanisms whereby lead is bound to humic and fulvic acids leads to the following conclusions: 1) because lead has a higher binding strength than other metals, lead can displace other metals on the organic molecule (Schnitzer, 1978); 2) if calcium is displaced, it would be leached to a lower soil horizon (B), where it may accumulate as it normally does during the development of the soil profile; and 3) if other nutrient metals, such as iron or manganese, are displaced, they may become unavailable to roots as they pass out of the soil system.

Fulvic acid plays an important role in the development of the soil profile. This organic acid has the ability to remove iron from the lattice structures of inorganic minerals, resulting in the decomposition of these minerals as a part of the weathering process. This breakdown releases nutrients for uptake by plant roots. If all binding sites on fulvic acid are occupied by lead, the role of fulvic acid in providing nutrients to plants will be circumvented. While it is reasonably certain that such a process is possible, there is no information about the soil lead concentrations that would cause such an effect.

Ecosystem inputs of lead by the atmospheric route have established new pathways and widened old ones. Insignificant amounts of lead are removed by surface runoff or ground water seepage. It is likely that the ultimate fate of atmospheric lead will be a gradual elevation in lead concentration of all reservoirs in the system, with most of the lead accumulating in the detrital reservoir.

8.6 SUMMARY

Because there is no protection from industrial lead once it enters the atmosphere, it is important to fully understand the effects of industrial lead emissions. Of the 450,000 tons emitted annually on a global basis, 115,000 tons of lead fall on terrestrial ecosystems. Evenly distributed, this would amount to 0.1 g/ha·yr, which is much lower than the range of 15-1,000,000 g/ha·yr reported in ecosystem studies in the United States. Lead has permeated these ecosystems and accumulated in the soil reservoir where it will remain for decades. Within 20 meters of every major highway, up to 10,000 µg Pb have been added to each gram of surface soil since 1930 (Getz et al., 1977c). Near smelters, mines, and in urban areas, as much as 130,000 µg/g have been observed in the upper 2.5 cm of soil (Jennett et al., 1977). At increasing distances up to 5 kilometers away from sources, the gradient of lead added since 1930 drops to less than 10 µg/g (Page and Ganje, 1970), and 1-5 µg/g have been added in regions more distant than 5 kilometers (Nriagu, 1978). In undisturbed ecosystems, atmospheric lead is retained by soil organic matter in the upper layer of soil surface. In cultivated soils, this lead is mixed with soil to a depth of 25 cm.

Because of the special nature of the soil reservoir, it must not be regarded as an infinite sink for lead. On the contrary, atmospheric lead that is already bound to soil will continue to pass into the grazing and detrital food chains until equilibrium is reached, whereupon the lead in all reservoirs will be elevated proportionately higher than natural background levels. This conclusion applies also to cultivated soils, where lead bound within the upper 25 cm is still within the root zone.

Few plants can survive at soil concentrations in excess of 20,000 µg/g, even under optimum conditions. Some key populations of soil microorganisms and invertebrates die off at 1000 µg/g. Herbivores, in addition to a normal diet from plant tissues, receive lead from the surfaces of vegetation in amounts that may be 10 times greater than from internal plant tissue. A diet of 2-8 mg/day·kg body weight seems to initiate physiological dysfunction in many vertebrates.

Whereas previous reports have focused on possible toxic effects of lead on plants, animals, and humans, it is essential to consider the degree of contamination as one measure of safe concentration. Observed toxic effects occur at environmental concentrations well above levels that cause no physiological dysfunction. Small animals in undisturbed ecosystems are contaminated by factors of 20-600 over natural background levels, and in roadside and urban ecosystems by 300-6200. Extrapolations based on sublethal effects may become reliable when these measurements can be made with controls free of contamination. The greatest impact may be on carnivorous animals, which generally have the lowest concentrations of natural lead, and may thus have the greatest percent increase when the final equilibrium is reached.

Perhaps the most subtle effect of lead is on ecosystems. The normal flow of energy through the decomposer food chain may be interrupted, the composition of communities may shift toward more lead-tolerant populations, and new biogeochemical pathways may be opened, as lead flows into and throughout the ecosystem. The ability of an ecosystem to compensate for atmospheric lead inputs, especially in the presence of other pollutants such as acid precipitation, depends not so much on factors of ecosystem recovery, but on undiscovered factors of ecosystem stability. Recovery implies that inputs of the perturbing pollutant have ceased and that the pollutant is being removed from the ecosystem. In the case of lead, the pollutant is not being eliminated from the system nor are the inputs ceasing. Terrestrial ecosystems will never return to their original, pristine levels of lead concentrations.

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